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GLOBAL CHANGE RESEARCH

Final Technical Report
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Water Pollution Impacts on Carbon Export and Greenhouse Gas Evasion from Asian River Systems

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Project Overview

Project Duration	:	3 years (15/07/2016–14/07/2019)
Funding Awarded	:	US\$ 43,000 for Year 1; US\$ 43,000 for Year 2; US\$ 43,000 for Year 3
Key organisations involved	:	<ol style="list-style-type: none">1. Ewha Womans University, Korea2. Royal University of Phnom Penh, Cambodia3. University of Washington, USA4. Physical Research Laboratory, India5. National University of Singapore, Singapore6. University of Hong Kong, China7. National Institute of Oceanography, India8. Jahangirnagar University, Bangladesh9. Cantho University, Vietnam10. Inner Mongolia University, China

Project Summary

Severe water pollution in rivers across Asia, resulting from rapid urbanization and poor wastewater management, has rarely been linked to carbon (C) export and greenhouse gas (GHG) emissions. This project aimed to build a research network focusing on C fluxes through Asian river systems to assess water pollution impacts on riverine GHG emissions. We took a unique approach combining synthesis workshops and exploratory field studies. Each year a workshop was held in a regional hub of the Mekong, Ganges, and Yellow River to synthesize monitoring data available from local sources and conduct a field trip followed by seasonal sampling surveys. Standardized monitoring protocols were disseminated among project members and research communities via a technical workshop session and a project webpage. Comparing literature information and local data with new field measurements suggests that rising organic pollution in rivers and tributaries downstream of metropolitan areas across the region has greatly increased organic C and GHG fluxes over the recent decades. These results call for concerted efforts among scientists and water agencies that can properly assess water pollution impacts on regional and global river carbon budgets and explore alternative policies for mitigating wastewater-induced increases in riverine GHG emissions.

Keywords: Asian rivers; carbon dioxide; greenhouse gases; river carbon; water pollution

Project outputs and outcomes

Project outputs:

- A. Scientific publications summarizing major findings on water pollution impacts on C export and GHG emissions from urbanizing river systems across Asia
 - A.1 Eight papers (literature reviews and research articles) published by project members in a special issue of an academic journal, Biogeosciences (https://www.biogeosciences.net/special_issue922.html)
 - A.2 Eight manuscripts (7 research articles and 1 synthesis) reporting findings from data syntheses and field studies in review or preparation

- B. Standardized carbon monitoring protocols disseminated via technical workshop sessions and the project homepage (<http://apn.peblab.com/>)
- C. A long-term research network that builds on the project members and invited speakers of the three project workshops
- D. Specific recommendations for alternative water policies integrating water quality control and climate change mitigation
 - D.1. Information about identified hot spots of riverine water pollution and GHG emissions that could help watershed managers set priorities for alternative water policies
 - D.2 A list of policy recommendations corresponding to demands of local and regional water agencies

Project outcomes:

- A. Contributing to scientific communities by providing new data and methods that are required to reduce uncertainties in estimating global river C budgets
- B. Capacity building of project members and other researchers in developing countries
- C. Providing a long-term collaboration framework for researchers studying river C fluxes across Asia
- D. Raising awareness of wastewater and polluted rivers as sources of GHGs among watershed managers and providing practical information for alternative policies integrating water quality control and climate change mitigation.

Key facts/figures

Key facts:

- New field measurements combined with literature and local monitoring data provided regional-scale estimates of riverine GHG emissions suggesting that Asian rivers have been underestimated as sources of GHGs, as illustrated by the new estimate of riverine CO₂ emission from the Indian Subcontinent (67 Tg C yr⁻¹) far exceeding the previous value (38 Tg C yr⁻¹).
- Extremely high levels of the partial pressure of CO₂ (pCO₂) measured in the Yamuna traversing Delhi (> 20,000 µatm) emphasize that rivers and tributaries downstream of metropolitan areas represent hotspots of riverine GHG emissions in rapidly urbanizing watersheds across Asia.

Key figures:

- Participants of the three workshops: 81 participants, including 28 in the first workshop, 20 in the second workshop, and 47 in the third workshop. In addition to 10 project members, 12 invited researchers presented works relevant to workshop topics.
- Students and researchers trained in the PI's lab: 1 postdoctoral researcher, 1 MSc researcher, 3 PhD students, 2 MSc. students
- Eight papers published by project members in a special issue of Biogeosciences and 8 manuscripts on key project findings in review or preparation

Potential for further work

Building on the collaboration network, including the project members and invited speakers of the three workshops, follow-up research activities can expand the duration of ongoing collaboration and the scope of addressed research topics. To add up to the first basin-wide field surveys of three GHGs along the Ganges, project members (Dr Kumar and Dr Tareq), in collaboration with PI, will continue field monitoring in India and Bangladesh, following

monitoring protocols established by this project. In situ incubation experiments and continuous sensor measurements of $p\text{CO}_2$, which were successfully implemented in the Mekong and Ganges, can also be applied to the Yellow River by project members (Drs. Lu, Ran and Yu) with technical support from PI. An ad-hoc review team will be formed to review emerging issues on altered river flow and C fluxes by newly constructed dams and aggravating water pollution in the Mekong River. While this project focused on the role of polluted urban rivers as sources of GHGs, a follow-up project can be envisaged to explore transitional stages of eutrophic rivers from a C sink, where phytoplanktonic CO_2 uptake exceeds CO_2 released from OM biodegradation to a source of GHGs, where emissions of CO_2 and other GHGs overwhelm temporary or seasonal increases in phytoplanktonic CO_2 uptake. These future activities adding to the project's focus on Asian river systems will contribute to elucidating human impacts on global riverine budgets of CO_2 and other GHGs.

Publications

- **Biogeosciences Special Issue: Human impacts on carbon fluxes in Asian river systems**
 - Editor(s): **J.-H. Park, V. V. S. S. Sarma**, G. Abril, and D. Butman
 - List of papers published by project members (project members indicated in bold font)
- Park, J.-H., Nayna, O. K., Begum, M. S.**, et al. (2018). Reviews and syntheses: Anthropogenic perturbations to carbon fluxes in Asian river systems – concepts, emerging trends, and research challenges. *Biogeosciences*, 15(9), 3049–3069. doi: 10.5194/bg-15-3049-2018
- Ran, L.**, Tian, M., Fang, N., Wang, S., **Lu, X.**, Yang, X., & Cho, F. (2018). Riverine carbon export in the arid to semiarid Wuding River catchment on the Chinese Loess Plateau. *Biogeosciences*, 15(12), 3857–3871. doi: 10.5194/bg-15-3857-2018
- Le, T. P. Q., Marchand, C., Ho, C. T., Duong, T. T., Nguyen, H. T. M., **Lu, X. X.**, ... Le, N. D. (2018). CO_2 partial pressure and CO_2 emissions from the lower Red River (Vietnam). *Biogeosciences*, 15, 4799–4814, doi: 10.5194/bg-15-4799-2018
- Dutta, M. K., **Kumar, S.**, Mukherjee, R., Sanyal, P., & Mukhopadhyay, S. K. (2019). The post-monsoon carbon biogeochemistry of the Hooghly–Sundarbans estuarine system under different levels of anthropogenic impacts. *Biogeosciences*, 16(2), 289–307. doi: 10.5194/bg-16-289-2019
- Lin, L., **Lu, X.**, Liu, S., Liang, S.-Y., & Fu, K. (2019). Physically controlled CO_2 effluxes from a reservoir surface in the upper Mekong River Basin: a case study in the Gongguoqiao Reservoir. *Biogeosciences*, 16(10), 2205–2219. doi: 10.5194/bg-16-2205-2019
- Jin, H., Yoon, T. K., **Begum, M. S.**, Lee, E.-J., Oh, N.-H., Kang, N., & **Park, J.-H.** (2018). Longitudinal discontinuities in riverine greenhouse gas dynamics generated by dams and urban wastewater. *Biogeosciences*, 15(20), 6349–6369. doi: 10.5194/bg-15-6349-2018
- Li, S., Mao, R., Ma, Y., & **Sarma, V. V. S. S.** (2019). Gas transfer velocities of CO_2 in subtropical monsoonal climate streams and small rivers. *Biogeosciences*, 16(3), 681–693. doi: 10.5194/bg-16-681-2019
- Krishna, M. S., Viswanadham, R., Prasad, M. H. K., Kumari, V. R., & **Sarma, V. V. S. S.** (2019). Export fluxes of dissolved inorganic carbon to the northern Indian Ocean from the Indian monsoonal rivers. *Biogeosciences*, 16(2), 505–519. doi: 10.5194/bg-16-505-2019



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Special issue

Human impacts on carbon fluxes in Asian river systems

Editor(s): J.-H. Park, V. V. S. S. Sarma, G. Abril, and D. Butman

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About

Editorial board

Awards and honours

Best poster award in the Korean Association of Biological Sciences Annual Meeting 2017.

- **Begum, M. S., Nayna, O. K., Jin, H., Lim, J., & Park, J.-H.** (2017). Longitudinal Variations in Dissolved Organic Matter and Greenhouse Gases in Three Asian River Systems. Korean Association of Biological Sciences Annual Meeting 2017. Seoul National University, Seoul, Republic of Korea.

Pull quote

“This workshop will also start the process of building community of experts and practitioners to effectively propagate and collaborate on carbon fluxes to assess water pollution impacts on Asian rivers as part of C sources. I am hoping that this regional collaboration will provide science-based policy recommendations on water management and climate change mitigation for sustainable water resource management in Asian region.” – from the welcome address (Third Workshop) by Dr Frank Yonghong Li (Dean of College of Ecology and Environment, Inner Mongolia University, China)

Acknowledgements

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1. Introduction

1.1. Backgrounds

Inland waters constitute a crucial component of the global carbon (C) cycle, playing many important roles including C storage in lake sediments and floodplains (Alin and Johnson, 2007), CO₂ exchange between the atmosphere and inland waters (Raymond et al., 2013), and riverine transports of dissolved organic C (DOC), particulate organic C (POC), and inorganic C (Battin et al., 2009). A growing body of evidence has shown that much more C is released from terrestrial sources to inland waters than previously thought and that inland waters function as an “active pipe” transforming C components during their transport to the oceans (Richey et al., 2002; Cole et al., 2007; Battin et al., 2009; Wehrli, 2013). Despite the importance of inland waters in the global C cycle, there remain considerable uncertainties about the amount and fate of anthropogenic C in inland waters (Regnier et al., 2013). Organic pollution and associated nutrient enrichment, combined with other anthropogenic changes such as river impoundment, significantly affect the riverine processing of organic matter (Stanley et al., 2012). Compared to anthropogenic C fluxes associated with erosion and weathering, much less is known about the characteristics and cascading effects of C contained in sewage and urban nonpoint-source runoff (Griffith et al., 2009; Butman et al., 2014). Although enhanced lability and mineralization of organic C have been measured in streams and rivers draining urbanized watersheds (Hosen et al., 2014; Kaushal et al., 2014), the chemical composition and lability of urban runoff and wastewater treatment plant (WWTP) effluents has rarely been associated with organic C mineralization and CO₂ evasion in large rivers and estuaries (Griffith and Raymond, 2011).

The information gap is particularly daunting when we address organic C transformations and CO₂ outgassing in streams and rivers draining rapidly urbanizing watersheds in developing countries across Asia. Rivers in many parts of Asia are highly polluted with domestic waste, agricultural runoff, and industrial waste, with the total volume of wastewater generated annually, excluding agricultural drainage, estimated at ~142 km³ (Evans et al., 2012). Polluted rivers and estuaries in Europe have been reported as a significant source of CO₂ and CH₄, with much of respired C coming from labile organic materials released from anthropogenic sources (Frankignoulle et al., 1998; Borges and Abril, 2011). Few efforts have been made to estimate greenhouse gas emissions from polluted Asian rivers using direct in-situ measurements of pCO₂, with a few exceptions for some large rivers and estuaries in China (Zhai et al., 2005; Chou et al., 2013; Ran et al., 2015). These studies, together with a small number of studies that used water chemistry data to estimate CO₂ evasion from major Asian rivers such as the Mekong (Li et al., 2013) and Indian estuaries (Sarma et al., 2012, 2013), pointed to the potential importance of anthropogenic organic matter and nutrients for seasonal dynamics of CO₂ evasion, particularly along rivers and estuaries downstream of densely populated areas. Direct underway measurements conducted in a few rivers and estuaries in China suggested some potential activation of riverine microbial processing and enhanced CO₂ evasion from polluted waterways (Zhai et al., 2005; Chou et al., 2013). Other studies have examined the effects of domestic and industrial wastewaters on the chemical composition and lability of riverine organic C (e.g., Guo et al., 2014), yet these attempts have not been linked to enhanced CO₂ evasion from polluted rivers and estuaries.

To better assess human impacts on riverine C fluxes, concerted efforts are required among experts across the region to generate more comprehensive data sets as well as direct in-situ measurements based on standardized monitoring techniques and protocols. Many of previous estimates of riverine CO₂ emissions were based on secondary water chemistry data including pH, alkalinity, and dissolved inorganic carbon (DIC) as inputs for modelling water-atmosphere gas delivery (e.g., Butman et al., 2012; Raymond et al., 2013). However, calculation of *p*CO₂ from pH and alkalinity can often result in large overestimation in freshwaters where acidity is strongly influenced by other factors than DIC chemistry, such as organic acids (Abril et al., 2015). While floating chambers or eddy flux towers can be deployed to measure the gas transfer between water and atmosphere in certain freshwater systems directly under favourable conditions, many technical challenges and/or high costs prevent the wide use of these techniques (Vachon et al., 2010). Several techniques have been used to measure *p*CO₂ in various inland water systems, including manual headspace equilibration using glass bottles or syringes (Hope et al., 1995), automated equilibrators (Frangkignoulle et al., 2001; Pierrot et al., 2009), and CO₂ sensors enclosed in gas-permeable membranes (Johnson et al., 2010). However, these techniques have rarely been compared for their accuracy and applicability to long-term field deployments. More field tests and standardization of procedural details are required to lower technical barriers for researchers in developing countries.

In sum, the rapid biological transformation of organic C in urbanized river systems and resulting enhancement of CO₂ outgassing represent not only an emerging research topic for environmental scientists but also a pressing challenge for government policies on water management and climate change mitigation. Scientifically rigorous assessments of human impacts on riverine C fluxes require building networks of researchers who can not only integrate available data and information but also provide field measurements of riverine C fluxes using standardized monitoring protocols.

1.2. Scientific significance

Despite the importance of inland waters in the global C cycle, considerable uncertainties exist with regard to the amount and fate of anthropogenic C in rapidly urbanizing river systems across Asia. Increasing emissions of CO₂ and other greenhouse gases (GHGs) from polluted rivers in Asia represent understudied sources of GHGs. Despite the quantitative importance of Asian rivers for global C fluxes, field-based studies have been very limited in both spatial coverage and employing standardized monitoring protocols.

The proposed project aims to provide the first regional assessment of water pollution impacts on C fluxes through major Asian rivers. Outcomes of the project can fill the information gap in our understanding of CO₂ emissions from streams and rivers draining rapidly urbanizing watersheds in developing countries across Asia. Combining new field measurements and literature data will also enlarge our understanding of the far-reaching impacts of wastewater on riverine metabolism and CO₂ emissions from large rivers in Asia.

1.3. Objectives

The primary objective of this regional collaborative research is building a collaborative research network to assess the current status of water pollution impacts on major Asian rivers as an underappreciated source of GHGs, using environmental monitoring data available from local sources and new measurements via exploratory field studies. Various

project activities and outcomes will be coordinated to build a long-term regional collaboration network focused on riverine biogeochemical cycles and GHG emissions. Project members with expertise in local environments and specific monitoring techniques, together with invited local collaborators, are involved in the literature review, data synthesis, and field studies, producing and sharing outcomes in the forms of standardized monitoring protocols, samples and data, and joint publications. These outcomes will be synthesized to produce the first regional assessment of water pollution impacts on the C fluxes through Asian rivers. In collaboration with local and regional water authorities and NGOs, scientific results will be translated into actionable information for water management options integrating water quality and GHG emissions and disseminated through the project homepage to raise public awareness of the land use-water-climate nexus.

Specific objectives are as follows:

- (1) The proposed research aims to bring together experts working on the C cycle in Asian river systems across the Asia-Pacific region to assess the current status of Asian rivers from the perspective of human impacts on riverine C transport and GHG evasion. Collaborative research will be implemented through interrelated project components:
 - Reviewing literature to provide an overview of the current status of water pollution and C fluxes in Asian river systems
 - Synthesis of available long-term monitoring data and new data from explorative field studies to evaluate the contribution of water pollution to riverine C fluxes in three regional hub basins
 - Contributing to capacity building of Asian researchers by establishing and disseminating standardized riverine C monitoring protocols
- (2) A policy-relevant objective is to provide local and regional policymakers and water managers with watershed management options integrating the conventional water quality focus and a new initiative for mitigating GHG emissions from natural ecosystems. Two specific goals have been set:
 - Providing science-based information to set priority areas and target ranges of GHG emissions required for proactive policymaking
 - Identifying policy demands and information paucity of local and regional water agencies and programs

2. Methodology

The project consisted of three interlinked components: (1) annual workshops with three sessions (literature review, data synthesis, and technical session) and a field trip testing and applying established monitoring protocols; (2) establishing and disseminating standardized C monitoring protocols; and (3) exploratory field studies comprising the workshop field trips and follow-up samplings (Figure 1). Results from three project components were synthesized to provide a regional-scale assessment of water pollution impacts on riverine CO₂ emissions. These results will form baseline data essential for identifying hot spots and moments of water pollution in river networks that result in enhanced riverine emissions of GHGs. The regional-scale assessment, along with watershed management options for reducing riverine emissions of GHGs, will help local water authorities and regional

organizations (e.g., Mekong River Commission) steer into next-generation watershed management policies incorporating both water quality control and GHG emission mitigation.

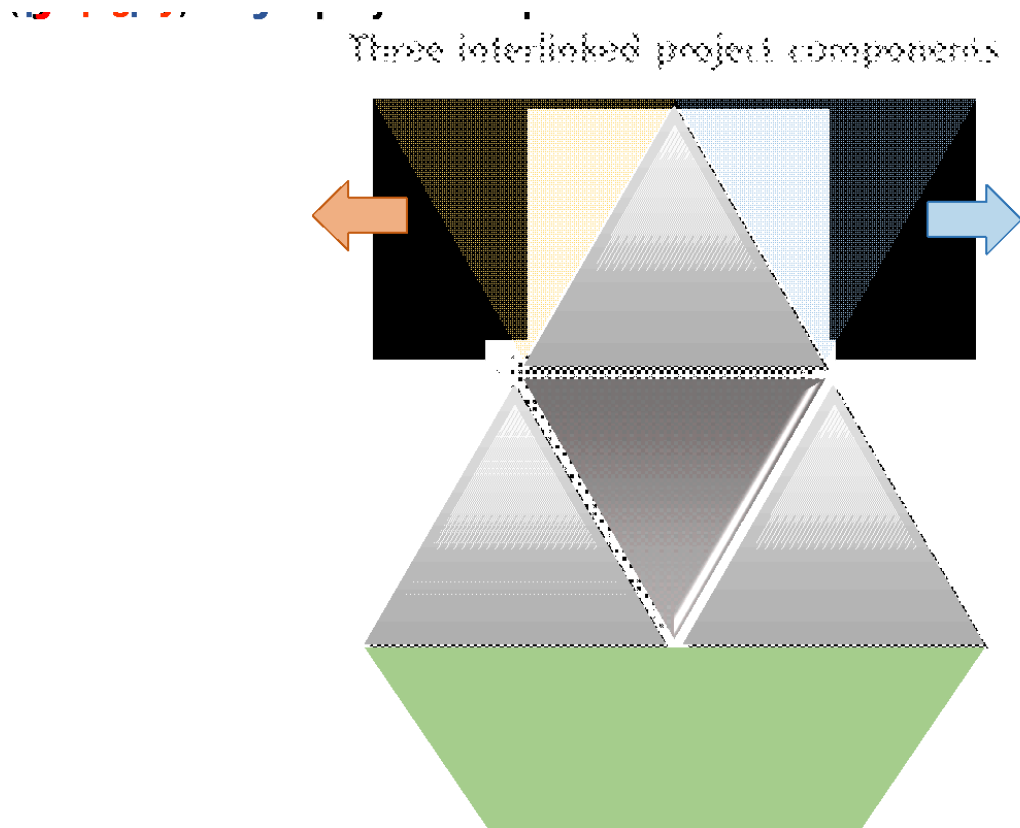


Figure 1. Overview of three interlinked project components and their specific goals

2.1. Annual workshops

Each year an annual workshop was held in each of three regional hubs – the Mekong, the Ganges-Brahmaputra, and Yellow River basins that represent East, Southeast, and South Asian regions, respectively. Main objectives of the workshops were: to obtain an overview of the current status and past studies on water pollution and its impacts on riverine C fluxes in each region; to synthesize long-term data available from local sources with an ultimate goal to produce regional-scale estimates of the riverine C export and GHG evasion; and to disseminate standardized C monitoring methods among project members and local collaborators via a technical session combined with a field trip.

The sessions were arranged for an in-depth discussion of emerging regional water issues confronting Asian countries and their implications for riverine C transport and transformations. These sessions also aimed at a synthesis of long-term environmental monitoring data such as discharge, water temperature, pH, alkalinity, and total suspended solid (TSS), with an ultimate goal to provide the most up-to-date estimates for regional-scale riverine exports of DOC and POC and emissions of CO₂ and CH₄ as well as the contributions of anthropogenic organic matter pollution to those C fluxes.

Historical records of relevant water quality parameters were retrieved from local sources (e.g., Yellow River Conservancy Commission) or global databases (e.g., GLORICH – Global River Chemistry Database). Routine water chemistry data collected by local sources were supplemented with direct measurements or calculations of DOC, POC, and pCO₂ that have

been collected by project members over scattered periods of time in the Yellow River (Ran et al., 2015), Mekong (Ellis et al., 2012; Li et al., 2013), and Ganges-Brahmaputra (Sarma et al., 2011, 2012). The partial pressure of CO₂ (pCO₂) was calculated using the CO2SYS program (Lewis et al., 1998), based on the speciation of the total DIC into HCO₃⁻, CO₃²⁻, H₂CO₃, and aqueous CO₂ depending on temperature and pH. The estimated fluxes of C species were compared to identify differences in C dynamics among different river systems and between up- and downstream reaches under different levels of anthropogenic perturbations. Each workshop included a technical session that provided project members and local collaborators with an overview of theoretical principles and applications of the standardized C monitoring techniques. All participants of the field trip could obtain hands-on experience in in-situ measurements of partial pressure of pCO₂ and water sampling for related water quality components (temperature, pH, conductivity, alkalinity, DOC, POC, ultraviolet absorption (UVA), and fluorescence excitation-emission matrices (EEMs), dissolved ions, and dissolved CH₄ and N₂O).

2.2. Establishing and disseminating standardized C monitoring protocols

A suite of laboratory and field experiments were performed at the project leader's laboratory in collaboration with other project members to establish low-cost, easy-to-operate C monitoring techniques. These protocols were tested in workshop field trips and follow-up samplings and then disseminated among project members and broader regional research communities. Widely used techniques for pCO₂ or CO₂ efflux measurements that are currently available at the laboratories of the project leader and other members, including headspace equilibration, CO₂ equilibrators, PTFE membrane-enclosed CO₂ sensors, and floating chambers, were compared against each other and the calculated pCO₂ values using the CO2SYS program for different water types under various environmental conditions (Figure 2). Building on the performance comparison results, step-by-step protocols were established for each of sensor deployment and in-situ measurements, data logging to analysis, and water sampling and analysis. Project members with experiences in CO₂ flux measurements provided feedbacks when the project leader circulated the protocol draft before test runs during the field trip.

While the main objective of the first year was to establish spatial survey methods, in the second year we focused on continuous pCO₂ monitoring techniques that can generate continuous measurements at finer temporal resolutions. The main goal of the last year was to explore "easy-to-use-everywhere" methods for simultaneous determination of three major GHGs – CO₂, CH₄, and N₂O – using GC-based analysis of headspace gas samples from air-tight sample containers. These standardized monitoring protocols were tested for field application during workshop field trips and follow-up samplings at selected study sites in each regional hub, also generating in-situ measurements of pCO₂ and key water quality components for joint publications. After the completion of field tests by project members, standardized monitoring protocols describing procedural details on instrumental set-ups, in situ measurements, and water sampling and analyses were posted on the project homepage for information dissemination among the broader research community.



Figure 2. Riverine C monitoring systems employed in field studies

2.3. Exploratory field studies on riverine C transport and GHG emissions

- **Study sites**

Explorative studies was focused on three large rivers originating from the Himalayas or the Tibetan Plateau, the world's largest (2,500,000 km²) and highest (average elevation >4000 m) plateau that holds the largest mass of ice on earth except the Antarctica and Greenland (Figure 3) (Spencer et al., 2014). The Ganges River and its headwater streams originate from the Himalayan region in India and Nepal, whereas the Mekong and Yellow rivers derive from the Tibetan Plateau in China. The two major branches of the upper Ganges called the Ganga locally and the Yamuna, join at the confluence near Varanasi and discharge to the Bay of Bengal through two major distributaries, namely the Hooghly in India and the Padma in Bangladesh, respectively. The longest stretch of the Ganges River from the origin of the Yamuna to the river mouth in Bangladesh is ~2950 km, draining a ~1.26×10⁶ km² watershed (Parua, 2009). The upper Ganges basin is covered by permafrost and large forested areas in the Himalayas under strong influence of the Indian summer monsoon, whereas the lower Ganges basin is influenced by deforestation, agricultural lands, livestock farms, wastewater discharge from the metropolitan areas, and 24 dams on the mainstem and tributaries (Whitehead et al., 2015; Indian Ministry of Water Resources, 2016).

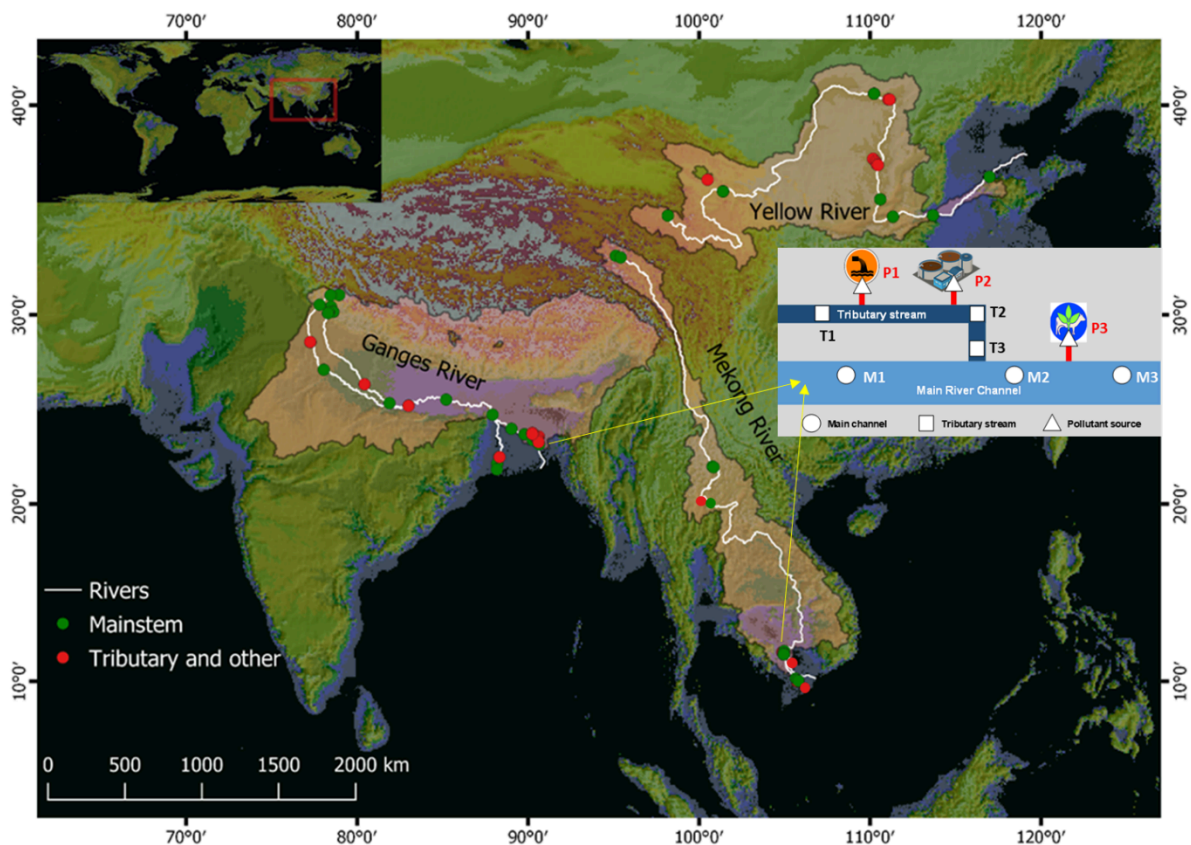


Figure 3. Sampling sites in three river systems (showing sites that have been sampled in July 2018; refer to Table 1 for more details on all sampling sites)

The Mekong River originates from the Tibetan Plateau in Qinghai Province, China. It flows through Myanmar, Laos, Thailand, Cambodia, and a complex delta system in Vietnam. The length of the Mekong River, the seventh longest river in Asia, is ~4800 km and drains an area of $800 \times 10^3 \text{ km}^2$ before emptying to the South China Sea (Milliman and Farnsworth, 2011). The upper Mekong river basin consists of mountainous terrain with an elevation between 400 and 5000 m and is affected by the melting snow from the Tibetan plateau (Li et al., 2013). A cascade of dams in the upper Mekong River basin, together with several ongoing dam construction projects on the mainstem and tributaries in the lower basin, might have significantly altered the hydrology and biogeochemical processes (Li et al., 2013). The lower Mekong River supports a population of about 70 million people causing much adverse effect from increasing level of fishing, agriculture, urbanization, and pollution (Li et al., 2013; Chea et al., 2016).

Yellow River also originates from a high elevation in the Tibetan Plateau in Qinghai Province and flows through nine provinces and discharges to the Bohai Sea in Shandong Province. The river is 5500 km long, draining an area of $750 \times 10^3 \text{ km}^2$ watershed including the Loess Plateau. The Yellow River has unique characteristics of a low discharge and a high sediment load from intense agriculture and livestock grazing on erosion-susceptible lands, although erosion-derived sediment yield has recently decreased owing to reforestation and construction of erosion dams across the major erosion source areas in the Yellow River basin (Milliman and Farnsworth, 2011).

Table 1. Summary of field samplings conducted in the Ganges-Brahmaputra, Mekong and Yellow River for 2016–2019. Samples were collected from the mainstem, local tributaries and wastewater drain feeding to the mainstem or tributaries.

River system	Reach	Site	Sampling points (number)	Sampling time	Note
Ganges–Brahmaputra	Upper	India	9	July–August 2017; February 2018	Headwater & upper reach; dry-wet season
	Middle	India	9	July–August 2017; February 2018; June 2019	Dry-wet season
	Lower	India-Bangladesh	24	August 2017; February 2018; February 2019	Dry-wet season
Mekong	Upper	Tsinghai, China	2	July 2016	Headwater; wet season
	Middle	Jinghong, China	2	August 2018; July 2019	Dry-Wet season
		Chiang Saen, Thailand	2	July 2019	Dry season
	Lower	Phnom Penh, Cambodia	11	January 2017; August 2017	Dry-wet season
Cantho, Vietnam		9	April 2017; August 2017	Dry-wet season	
Yellow River	Upper	Tsinghai, China	2	July 2016	Headwater & upper reach; wet season
	Middle	Hohhot, Hancheng, China	3	September 2017;	Dry-wet season
			2	May/August 2018	
Lower	Zhengzhou, Jinan, China	1	May 2018	Dry season	

- **Water and dissolved gas sample collection**

Two-season samplings were conducted in the three basins during a pre-monsoon (dry period) and a monsoon (high-flow period) season between 2016 and 2019, although the number of sampling sites differed in each sampling campaign. However, one-time sampling was conducted at most of the Yellow river sites due to logistical constraint. Grab water samples together with dissolved gas samples were collected from 10–20 cm below the water surface at 42 sites in the Ganges, 26 sites in the Mekong and 9 sites in the Yellow River systems. In addition to collecting samples from the mainstem, urban tributaries and wastewater drain feeding to the mainstem or tributaries were also collected in order to examine the effect of urbanization and water pollution on large Asian rivers. PI's laboratory collected samples and/or project members and analysis was conducted at Ewha Womans University following the standard protocols developed by the PI's laboratory. In-situ measurements included water quality parameters such as water temperature, pH, dissolved oxygen (DO), and electrical conductivity (EC) as well as barometric pressure and air

temperature. Samples were transported on ice and kept frozen before laboratory analysis at Ewha Womans University.

Dissolved gas samples were collected by manual headspace equilibration method (Yoon et al., 2016). Headspace equilibration was performed using a 60 mL polypropylene syringe to collect a 30 mL water sample and then a 30 mL ambient air sample. Another air sample was collected to measure gas concentrations in the ambient air. The syringe containing the water and air samples was shaken vigorously for 2 min; then ~20mL of the equilibrated air was stored in a pre-evacuated 12 mL Exetainer vial. Gas analysis was performed within a month after the sampling. Barometric pressure and water temperature were used to calculate concentrations of the three GHGs from the gas concentrations of the equilibrated air and ambient air samples based on Henry's law (Hudson, 2004).

- Water and gas analysis

Water samples were filtered through pre-combusted glass fibre (GF/F) filters to remove suspended materials. The concentration of DOC in the filtered water sample was measured using a total organic carbon (TOC) analyser based on high-temperature combustion of organic matter (OM) followed by thermal detection of CO₂ (TOC-V_{C_{PH}}, Shimadzu, Japan). Filtered water samples were also analysed for major inorganic ions including NO₃⁻ and PO₄³⁻ (883 Basic IC Plus, Metrohm, Switzerland). Total Alkalinity (TA) was measured with 40–80 mL filtered samples on an automated electric titrator (EasyPlus Titrator Easy pH, Metrohm, Switzerland) based on the Gran titration method.

UVA was measured across the wavelength range from 200 to 1100 nm using a UV-Vis spectrophotometer, and specific UVA at 254 nm (SUVA₂₅₄) was determined from UV-vis absorbance measurements (Weishaar et al., 2003; Helms et al., 2008). Fluorescence EEMs were collected on a fluorescence spectrophotometer (F7000, Hitachi, Japan) by simultaneous scanning overexcitation wavelengths from 200 to 400 nm at 5 nm interval and emission wavelengths from 290 to 540 nm at 1 nm interval, as detailed in Begum et al. (2019). Fluorescence-based indices were obtained from the corrected EEMs, including humification index (HIX) (Zsolnay et al., 1999), fluorescence index (FI) (McKnight et al., 2001), and biological index (BIX) (Huguet et al., 2009), following standard methods from the references. As part of quality control, standards with known concentrations and ultrapure water were analysed for every batch of ten samples, and triplicate analysis was performed for approximately 10% of all the samples to assess instrumental stability and accuracy.

Fluorescent dissolved organic matter (FDOM) components were identified from corrected EEMs by parallel factor analysis (PARAFAC) using MATLAB 7.1 (Mathworks, Natick, USA) and the DOM Fluor toolbox (<http://www.models.life.ku.dk>) based on a method developed by Stedmon and Bro (2008). The maximum fluorescence intensities (F_{max}) of identified components were used to represent their absolute and relative abundance (%) in each sample. FDOM components were termed based on the similarity of the peak wavelengths to values reported in the literature (Fellman et al., 2010).

Solid-phase extraction (SPE) was used to extract dissolved organic matter (DOM) using a sorbent made of styrene-divinylbenzene (bond elute PPL) and methanol as eluent, as described in detail in Begum et al. (2019). Samples were analysed in 15 Tesla Fourier transform-ion cyclotron resonance (FT-ICR) mass spectrometer (Bruker Daltonics Inc., Germany) equipped with an Apollo II ion funnel electrospray ionization (ESI) source, at Korea Basic Science Institute (KBSI, Ochang, South Korea). Molecular formula assignments

were made based on several general guidelines suggested by Koch et al. (2007) and figures were plotted using Matlab or SigmaPlot.

The equilibrated headspace air sample, as well as an ambient air sample used for equilibration, was measured on a gas chromatography (GC) (7890A, Agilent, USA) equipped with an FID coupled with a methanizer (for analysis of CH₄ and CO₂), a μ ECD (for N₂O analysis) for simultaneous measurement of three GHGs. Another set of samples were analysed for stable C isotope ratios of CO₂ and CH₄ by an isotopic analyser (G2201-i, Picarro, USA) in the laboratory of David Butman, University of Washington.

- **Continuous underway measurement of $p\text{CO}_2$ in the Mekong and Ganges**

In addition to point samplings, continuous underway measurement of $p\text{CO}_2$ were performed along the Mekong-Tonle Sap around Phnom Penh, Cambodia (January 2017) and Buriganga-Meghna-Ganges near Dhaka, Bangladesh (February 2018) as part of the field works during the first and second APN workshops. The cruise expeditions were conducted along the river and a local urban tributary in order to explore the effect of urban wastewater on spatial variations in water quality and $p\text{CO}_2$. Continuous, in-situ measurements of $p\text{CO}_2$ were conducted using a membrane-enclosed CO₂ sensor (GMT222, Vaisala, Finland) connected to a data logger (CR10X; CR1000, Campbell Scientific Inc., USA) and a spray-type equilibrator connected to an infrared gas analyser (IRGA; LI820, Li-Cor), as detailed in Yoon et al. (2016). Basic water quality parameters, including water temperature, atmospheric pressure, pH, electrical conductivity and DO, were also measured simultaneously during the cruise expedition using the portable multiparameter meter (6820 V2, YSI Inc.).

- **In-situ incubation experiments**

In order to examine the effect of wastewater on DOM biodegradability under field conditions, in-situ incubation experiments were conducted at a downstream location of the Mekong near Phnom Penh and the lower Ganges near Dhaka during the first and second workshop. Water samples from a mainstem site and a wastewater drain, together with a mixture of the mainstem and the sewage sample in 9:1 ratio, were incubated for 3 days with and without light exposure. Continuous $p\text{CO}_2$ measurement was recorded in the 1st incubation experiment in the Mekong, while dissolved CO₂ was measured before and after the incubation in the case of 2nd incubation experiment in the Ganges. The 3-day incubation was conducted in transparent bottles to measure the net biodegradable dissolved organic carbon (BDOC) under the field condition. Another set of samples were incubated in opaque (covered with aluminium foil) bottles under the same condition to measure total BDOC without influence of photodegradation and autochthonous production. SPE-DOM samples from Ganges were also analysed using FT-ICR-MS before and after the incubation in order to detect molecular-level changes in DOM composition during the incubation.

- **Monthly monitoring of $p\text{CO}_2$ in the lower Ganges, Bangladesh**

As a follow-up activity of the field campaign during the second workshop in Dhaka, longer-term monitoring of $p\text{CO}_2$ and basic water quality parameters was conducted in a Ganges location near Hardinge Bridge, Kushtia, Bangladesh by the research group of a project member (Dr Tareq). Water and dissolved gas samples were collected from the same site every month from May 2018 to April 2019. In this long-term monitoring, membrane-enclosed CO₂ sensors and a water quality probe were deployed for 12 hours during their monthly sampling and data was continuously recorded in a logger at 10-second

interval in order to measure diurnal variations in surface water $p\text{CO}_2$, together with water temperature, pH, EC, and DO.

2.4. Synthesis and science-policy translation

Results from data synthesis workshops and exploratory field studies were compiled into a database like core data for up-scaling to significant regions of Asia to estimate regional C fluxes and contributions of water pollution to those fluxes. To take advantage of existing global data sets on river flow and basic water chemistry, we used GLORICH (Global River Chemistry Database) for pH and alkalinity data required for $p\text{CO}_2$ calculations, CPCB (Central Pollution Control Board, India) and published literature for BOD data. Spatially more resolved data obtained from field surveys and local sources were compared with literature data to improve the resolution of the existing global data, allowing us to obtain better estimates of anthropogenic C loadings and their contributions to riverine C fluxes.

A policy-relevant project goal was to provide policymakers and the general public with science-based alternative watershed management options integrating climate change mitigation in the water sector. The collaboration with local research collaborators and government officials allowed us to identify key issues and priorities for water management in the studied river basins. In the case of Cambodia and Bangladesh where the first two workshops were held, the lack of wastewater treatment facilities and associated river pollution represented the foremost concern and policy priority for local water authorities. Downstream ecosystem impacts and GHG emissions have not been incorporated into any pro-active policy alternatives. To compare the poor wastewater infrastructure in the Mekong and Ganges basins with growing environmental awareness and technological development in China, the third workshop included a discussion session and interviews with local watershed managers of the Yellow River Conservancy Commission (YRCC).

Based on the regional assessment identifying hot spots and moments of anthropogenic C fluxes, priority areas demanding immediate policy actions will be recommended, along with predictions for climate change mitigation resulting from abated anthropogenic C loadings to the “hot spot” river reaches. An ultimate goal of our project is to provide policymakers and the general public with science-based alternative watershed management options integrating climate change mitigation in the natural resources and ecosystems sector. To facilitate communication with government officials and general public across Asia, project findings and policy recommendations will be summarized in newsletters and policy briefs for dissemination to the government offices and NGOs with which project members have collaborated throughout the project.

- **Estimating $p\text{CO}_2$ using an aquatic C equilibrium model (CO2SYS)**

Among various direct and indirect approaches used for determining $p\text{CO}_2$ in inland waters, the headspace equilibration method has widely been used as a direct $p\text{CO}_2$ measurement method. The water-air difference in $p\text{CO}_2$ was determined from the $p\text{CO}_2$ measurements in the equilibrated air and ambient air of the samples, later incorporated into a gas transfer model (Liss and Slaster., 1974, Deacon., 1977, Wanninkhof, 1992, Raymond and Cole., 2001, Wanninkhof et al., 2009). The equilibrated air and ambient air CO_2 concentrations were used together with water temperature and barometric pressure to calculate $p\text{CO}_2$ based on Henry’s law (Hudson, 2004).

Values of $p\text{CO}_2$ measured by headspace equilibration technique were compared with the calculated values using CO2SYS program to improve the prediction capability of the $p\text{CO}_2$

model for a regional synthesis based on water quality data available from local sources. The scarcity of direct measurements of $p\text{CO}_2$ at regional and global scales has resulted in obtaining $p\text{CO}_2$ data that was calculated using the CO2SYS program (Pierrot et al., 2006) which has widely been used to calculate riverine $p\text{CO}_2$ from temperature, pH and total alkalinity in estimating riverine CO_2 outgassing.

- **Estimating TOC and GHGs using BOD data**

DOC and TOC were calculated from actual BOD measurements, using some empirical equations such as $BOD = X \times DOC$ (X = conversion factor), $BOD = X \times TOC$ (X = conversion factor). River-specific conversion factors based on water types (mainstem, tributary and wastewater) obtained from different published literature. Actual measurements of BOD along the upper, middle and lower reaches of Ganges obtained from CPCB (Central Pollution Control Board) and literature in 1986–2014 used to calculate TOC and DOC using BOD_5/TOC and BOD_5/DOC ratios from different published literature respectively. DOC/CO_2 ratio obtained from 2017–2018 field data along the Ganges used to calculate BOD based CO_2 (Figure 4). BOD based empirical approach used to calculate the riverine DOC and CO_2 along the downstream of the rivers which are not included in the IPCC methods (Doorn et al., 2006).

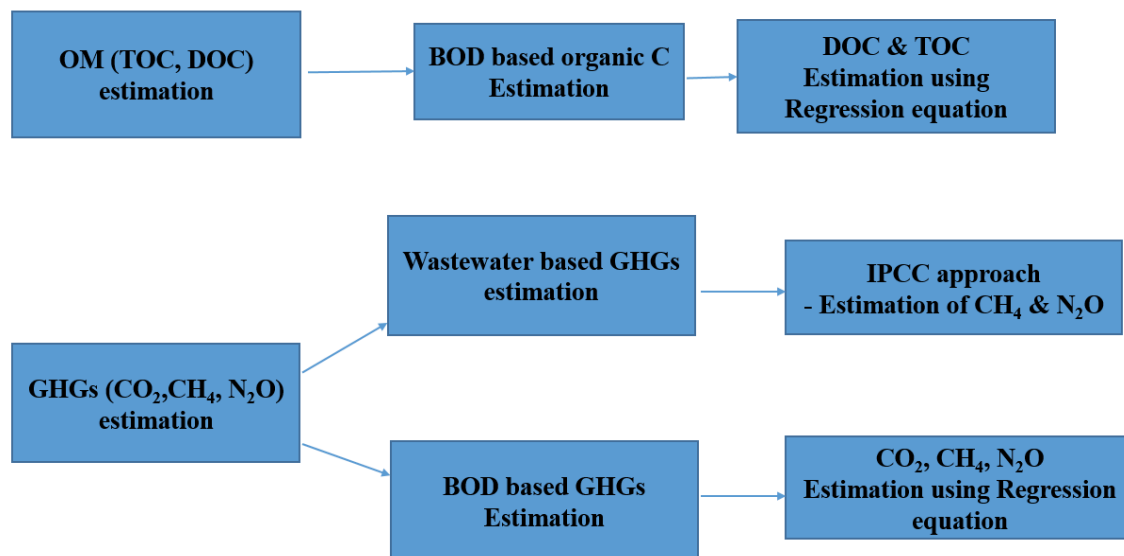


Figure 4. Methods to estimate OM and GHGs based on wastewater based IPCC approach and BOD based empirical approach.

IPCC approach was used to estimate wastewater (domestic and industrial wastewater) based GHGs (CH_4 and N_2O). Firstly, the total organic in domestic wastewater (TOW) was calculated using the given equation:

$$TOW = P \times BOD \times 0.001 \times I \times 365$$

Where, TOW = total organics in wastewater in inventory year, kg BOD per year; P = Country population in inventory year, (person); BOD = Country-specific per capita BOD in inventory year, g per person per day; 0.001 = Conversion from grams BOD to kg BOD; I = Correction factor for additional industrial BOD discharged into sewers. Total degradable organic material in industrial wastewater (TOW_i) was also calculated using the following equation:

$$TOW_i = P_i \times W_i \times COD$$

Here, TOW_i = total organically degradable material in wastewater for industry i , kg COD per year; i = industrial sector; P_i = total industrial product for industrial sector i , t per year; W_i = wastewater generated, m³/t product; COD_i = chemical oxygen demand, kg COD per m³.

Total CH₄ emissions from domestic wastewater were calculated using the given equation:

$$CH_4 \text{ Emission} = \sum_{ij} (U_i \times T_{ij} \times EF_j) (TOW - S) - R$$

Here, CH₄ Emissions = CH₄ emissions in inventory year, kg CH₄ per year; TOW = total organics in wastewater in inventory year, kg BOD per year; S = organic component removed as sludge in inventory year, kg BOD per year; U_i = fraction of population in income group i in inventory year; $T_{i,j}$ = degree of utilization of treatment/discharge pathway or system, j , for each income group fraction i in inventory year; i = income group: rural, urban high income and urban low income; j = each treatment/discharge pathway or system; EF_j = emission factor, kg CH₄ / kg BOD; R = amount of CH₄ recovered in inventory year, kg CH₄ per yr.

Total CH₄ emissions from industrial wastewater were calculated using the given equation:

$$CH_4 \text{ Emission} = \sum_i [(TOW_i - S_i) EF_i - R_i]$$

Here, CH₄ Emissions = CH₄ emissions in inventory year, kg CH₄ per year; TOW_i = total organically degradable material in wastewater from industry i in inventory year, kg COD per year; i = industrial sector; S_i = organic component removed as sludge in inventory year, kg COD per year; EF_i = emission factor for industry i , kg CH₄ per kg COD for treatment/discharge pathway or system(s) used in inventory year; R_i = amount of CH₄ recovered in inventory year, kg CH₄ per year. Wastewater effluent based N₂O emissions were calculated using the given equation:

$$N_2O \text{ Emission} = N_{EFFLUENT} \times EF_{EFFLUENT} \times 44/28$$

Where, N₂O emissions = N₂O emissions in inventory year, kg N₂O per year; $N_{EFFLUENT}$ = nitrogen in the effluent discharged to aquatic environments, kg N per year; $EF_{EFFLUENT}$ = emission factor for N₂O emissions from discharged to wastewater, kg N₂O-N/kg N. The factor 44/28 is the conversion of kg N₂O-N into kg N₂O.

- **Regional synthesis – The case of the Indian subcontinent**

Water quality data (pH, temperature and alkalinity) of major rivers in the Indian subcontinent, including the Ganges, Brahmaputra, Indus, Narmada, Krishna, Godavari and Mahanadi, were compiled from the literature. The compiled data provides comprehensive spatial representation for all rivers in the region, with the included rivers accounting for ~90% of total river discharge. The CO₂SY program was used to calculate pCO_2 based on pH, alkalinity and water temperature (Pierrot et al., 2006).

Water-air CO₂ exchange

Calculated values of pCO_2 were used to estimate water-air CO₂ flux using a gas evasion model given below,

$$F = A \times K_h \times (pCO_{2 \text{ water}} - pCO_{2 \text{ air}})$$

Here, the ambient air CO₂ ($pCO_{2 \text{ air}}$ in μatm) is subtracted from the surface water partial pressure of CO₂ ($pCO_{2 \text{ water}}$ in μatm) and then the difference is multiplied by A (river surface area) and K_h (the gas transfer velocity of CO₂ in cm/h). K_h can be calculated using the two values of streamflow velocity v and the channel slope S_{chan} using the equation from Raymond et al. (2012),

$$ms^{-1}) = -1.64 + 0.285 \times \ln(Q_{ann}) [m^3 s^{-1}] \text{ (after Raymond et al. 2012)}$$

$$ms^{-1}) = -1.06 + 0.12 \times \ln(Q_{ann}) [m^3 s^{-1}] \text{ (after Raymond et al. 2013)}$$

$$k_h [md^{-1}] = V [ms^{-1}] \times S_{chan}[1] \times 2841 + 2.02$$

3. Results & Discussion

3.1. Annual workshops

The first workshop was held at Royal University of Phnom Penh, Cambodia, for three days (January 11 – 13, 2017) (Table 2). The primary objective of the first workshop was to provide a venue for an in-depth discussion of emerging regional water issues confronting Asian countries and their implications for riverine C transport and transformations. The main workshop sessions included the project leader's introduction to the project, two invited talks (David Butman and Gordon Holtgrieve, University of Washington), and three sessions on the Mekong, Ganges-Brahmaputra, and Yellow River. Local organizer (Eliyan Chea, Royal University of Phnom Penh) invited researchers and government officers to disseminate workshop outcomes (Table 2).

The second workshop was held at Dhaka, Bangladesh for three days (February 9 – 11, 2018) (Table 3). The specific objectives of the second workshop were to obtain an overview of the current status of water pollution and its impacts on riverine C fluxes in the Ganges and other focal river basins; to find out long-term data available from local sources with an ultimate goal to produce regional-scale estimates of the riverine C export and GHG emissions; and to disseminate standardized C monitoring methods among project members and other workshop participants through a technical session combined with a field trip. The main workshop sessions included the project leader's introduction to the project, an invited talk (Tarun T. Dalai, Indian Institute of Science Education and Research Kolkata), and three sessions on the Mekong, Ganges-Brahmaputra, and Yellow River. Local organizer (Shafi M. Tareq, Jahangirnagar University) invited students and researchers to disseminate workshop outcomes.

The third workshop was held in Hohhot, China on 11 – 13 May 2019 (Table 4), with a regional focus on the Yellow River in addition to the objectives set for the three annual workshops. Main objectives of the annual workshops include: to obtain an overview of the current status and past studies on water pollution and its impacts on riverine C fluxes in each region; to synthesize long-term data available from local sources with an ultimate goal to produce regional-scale estimates of the riverine C export and GHG evasion; and to disseminate standardized C monitoring methods among project members and local collaborators via a technical session combined with a field trip. Building on the outcomes of the previous two workshops, the last workshop focused on discussions of how to synthesize new field data from this project and secondary data from local monitoring data sources and global databases (e.g., AQUASTAT, GEMS/Water, and GLORICH) to produce input data for a regional-scale assessment of pollution impacts on river water quality in general and levels of GHGs in particular. Project members presented their assessment of the local river status focusing on water pollution impacts on riverine C fluxes.

In all workshops, a special session on carbon flux monitoring was arranged to provide project members and local collaborators with an overview of theoretical principles and applications of the standardized C monitoring techniques that were established as part of the second project component. All participants of the field trip obtained hands-on experience in in-situ measurements of partial pressure of CO₂ (pCO₂) and water sampling for related water

quality components (temperature, pH, conductivity, alkalinity, DOC, POC, UVA, and fluorescence excitation-emission matrices, dissolved ions, and dissolved CH₄ and N₂O).

Table 2. List of the participants of the first workshop at Phnom Penh, Cambodia

Name	Affiliation	Country	Presentation Title
Invited Speakers			
David Butman	University of Washington	USA	A national-scale assessment of carbon fluxes from inland waters of the US in the context of the 2 nd U.S. State of the Carbon Cycle Report
Gordon Holtgrieve	University of Washington	USA	Flood-pulse controls on carbon fluxes and fishery productivity in the Mekong River and Tonle Sap Lake
Project Members			
Ji-Hyung Park	Ewha Womans University	Republic of Korea	Water pollution impacts on carbon export and greenhouse gas evasion from Asian river systems – Project overview
Xi Xi Lu	National University of Singapore	Singapore	Carbon outgassing from the Yellow River: An overview
Lishan Ran	University of Hong Kong	China	Riverine CO ₂ emissions from the Wuding River catchment on the Loess Plateau: Environmental controls and dam impoundment impact
Ruihong Yu	Inner Mongolia University	China	Spatio-temporal variations and influencing factors of carbon dioxide evasion from the Yellow River: an example of the Toudaoguai Station
VVSS Sarma	CSIR- National Institute of Oceanography	India	Trace gases emissions from the Indian estuaries
Shafi M. Tareq	Jahangirnagar University	Bangladesh	Using 3DEEM Fluorescence Spectroscopy for Monitoring Water Quality and DOM Dynamics in the Ganges and Brahmaputra River in Bangladesh
Jeffrey E. Richey	University of Washington	USA	Seasonal dynamics of the sources and isotopic composition of dissolved inorganic carbon in large tropical river systems: the Mekong and Chao Phraya rivers of Southeast Asia
Eliyan Chea	Royal University of Phnom Penh	Cambodia	Current Water Quality at Mekong River
Do Thi Xuan	Can Tho University	Vietnam	Dynamic Changes of Quality of Surface Water in the Mekong River in Vietnam from 2013-2015
Tae Kyung Yoon	Ewha Womans University	Republic of Korea	Water-air CO ₂ Flux Measurement Methods: Principles, Validations, and Challenges
Most Shirina Begum	Ewha Womans University	Republic of Korea	Sampling and analysis protocols for assessing human impacts on the riverine carbon fluxes in Asian river systems

Table 3. List of the participants of the second workshop at Dhaka, Bangladesh

Name	Affiliation	Country	Presentation Title
Invited Speaker			
Tarun K. Dalai	Indian Institute of Science Education and Research Kolkata	India	Enhanced delivery of dissolved inorganic carbon from the Ganga (Hooghly) River estuary to the Bay of Bengal: Sources, processes and fluxes
Project Members			
Ji-Hyung Park	Ewha Womans University	Republic of Korea	Assessing pollution impacts on carbon fluxes in Asian river systems – Workshop outcomes, exploratory field studies, and data synthesis
VVSS Sarma	CSIR- National Institute of Oceanography	India	Export fluxes of dissolved inorganic carbon to the Northern Indian Ocean from the Indian monsoonal rivers
Sanjeev Kumar	Physical Research Laboratory	India	Effect of a tidal cycle on biogeochemistry of mangrove dominated tropical estuary (Sundarbans, India)
Shafi M. Tareq	Jahangirnagar University	Bangladesh	Characteristic and Dynamic of Dissolved Organic Matter in the Ganges and Brahmaputra River, Bangladesh
Xi Xi Lu	National University of Singapore	Singapore	Carbon emission from Tibet Plateau rivers: A case study of the Yellow River headwater region
Eliyan Chea	Royal University of Phnom Penh	Cambodia	Phnom Penh Wastewater Management and Pressures to River Water Quality
Most Shirina Begum	Ewha Womans University	Republic of Korea	Organic matter and greenhouse gas dynamics in three Asian River systems - monitoring protocol and preliminary results

Table 4. List of the participants of the third workshop at Hohhot, China

Name	Affiliation	Country	Presentation Title
Invited Speaker			
Andreas Lorke	University of Koblenz, Landau	Germany	Spatial and temporal variability of methane emission from cascading reservoirs at the Upper Mekong River
Zaihua Liu	Chinese Academy of Sciences	China	Carbon sink by coupled carbonate weathering with aquatic photosynthesis—Control of climate and land-use changes
Qiuwen Chen	Nanjing Hydraulic Research Institute	China	Spatial patterns of GHG flux in the cascaded reservoirs along Lancang River and carbon-nitrogen biogeological cycles in the reservoir islands
Mingming Hu	China Institute of Water Resources and Hydropower Research	China	Export fluxes of dissolved inorganic carbon to the Northern Indian Ocean from the Indian monsoonal rivers
Haizhu Hu	Inner Mongolia University	China	Carbon dioxide emission from surface water in cascade reservoirs of the Yellow River source region
Project Members			
Ji-Hyung Park	Ewha Womans University	Republic of Korea	Synthesis of Project Outcomes toward a Regional-Scale Assessment of Water Pollution Impacts on Greenhouse Gas Emissions from Asian River Systems
Sanjeev Kumar	Physical Research Laboratory	India	Nitrogen and carbon cycling study in terrestrial aquatic and forested ecosystems in India
Shafi M. Tareq	Jahangirnagar University	Bangladesh	Investigating water quality of the Ganges and Brahmaputra river using three-dimensional excitation-emission matrix (3DEEM) fluorescence spectroscopy
Xi Xi Lu	National University of Singapore	Singapore	Carbon emissions from China's water bodies: an overview
Lishan Ran	University of Hong Kong	China	Hydrological controls on riverine export of water, sediment, and carbon: a case study of the Wuding River on the Chinese Loess Plateau
Ruihong Yu	Inner Mongolia University	China	Temporal and spatial variation of major ion chemistry and dissolved carbon in the source region of the Yellow River
Most S. Begum	Ewha Womans University	China	Assessing Greenhouse Gas and Organic Matter Flux and Quality in Urbanized Asian Rivers
Omme K. Nayna	Ewha Womans University	Republic of Korea	Three approaches to assessing pollution impacts on riverine organic carbon load and greenhouse gas emissions
Other Participants			
Yuchun Wang		China	China Institute of Water Resources and Hydropower Research
Shanfeng Zhao		China	Ningmeng Bureau of Hydrology and Water Resources, YRCC
XinWei Guo		China	Ningmeng Bureau of Hydrology and Water Resources, YRCC

3.2. Monitoring protocols

Monitoring protocols for measuring riverine carbon fluxes established in the first year have been improved through the laboratory and field experiments conducted by the project leader's research group. These monitoring methods have been tested and disseminated among the workshop participants during the workshop field trips in Phnom Penh in the first year and Dhaka in the second year (Figure 5). A revised version of the monitoring protocols in 25 pages had been prepared by incorporating feedback from the project members before it was made accessible to the public through the project webpage (<http://apn.peblab.com>) to disseminate the protocols among project members and broader research communities.

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Figure 5. Monitoring protocols (contents)

3.3. Field surveys and other exploratory field work in three river systems

3.3.1. Inter-basin comparison of water quality and GHGs

Concentrations of dissolved GHGs were highly variable in all three river systems (Table 5). Levels of $p\text{CO}_2$ varied across two orders of magnitude, ranging between 180–20580 μatm , 381–3729 μatm , and 611–4861 μatm in the Ganges, the Mekong, and the Yellow River, respectively. The highest average $p\text{CO}_2$ was found in the Mekong, followed by the Ganges and Yellow River. Variations in dissolved CH_4 concentrations spanned up to five orders of magnitude, ranging between 2–172132 nmol L^{-1} , 9–1893 nmol L^{-1} , and 59–1727 nmol L^{-1} in the Ganges, the Mekong, and the Yellow River, respectively. While the Mekong had the highest mean concentration of CH_4 , the lowest mean concentration of CH_4 was found in the Yellow River. Dissolved N_2O concentrations exhibited relatively small variations, ranging between 2–245 nmol L^{-1} , 3–24 nmol L^{-1} , and 8–63 nmol L^{-1} in the Ganges, the Mekong, and the Yellow River, respectively. Unlike the other GHGs, the average value was in descending order of the Ganges, the Yellow River, and the Mekong.

Table 5. Summary of the concentration of three GHGs, DOC, and POC in the Ganges, Mekong and Yellow River. Average values are followed by ranges in parentheses.

Sample	$p\text{CO}_2$ (μatm)	CH_4 (nmol L^{-1})	N_2O (nmol L^{-1})	DOC (mg L^{-1})	POC (mg L^{-1})
Ganges	4606 (180–27290)	24403 (2–234820)	58.0 (0.5–1699.9)	4.5 (0.2–31.3)	6.2 (0.5–39.5)
Mekong	5253 (381–30221)	25339 (9–455754)	20.1 (2.7–197.9)	3.9 (0.5–50.2)	3.5 (0.6–36.7)
Yellow	1910 (611–4861)	603 (60–3754)	40.0 (7.6–143.8)	5.1 (1.4–11.4)	5.0 (0.7–15.8)

Most sampling sites exhibited supersaturation of CO_2 relative to the level equivalent to atmospheric equilibrium, except a few headwater and mainstem sites in the upper reaches of the Ganges and the Mekong. CH_4 also showed supersaturation at all the sites except two upstream sites in the Ganges, whereas N_2O values oscillated between under- and supersaturation in all three rivers. Similar to the trends in three GHGs, DOC concentration also exhibited large variations in the three rivers (Table 5). DOC concentration spanned up to three orders of magnitude, ranging between 0.19–26.27 mg L^{-1} , 0.52–3.75 mg L^{-1} and 1.37–10.37 mg L^{-1} in the Ganges, the Mekong, and the Yellow River, respectively. High values of DOC were usually found in the urban tributaries. POC concentration also showed large variations with values ranging between 0.5–39.5 mg L^{-1} , 0.6–36.7 mg L^{-1} , and 0.7–15.8 mg L^{-1} in the Ganges, the Mekong, and the Yellow River, respectively. Highest values of POC were observed in wastewater samples.

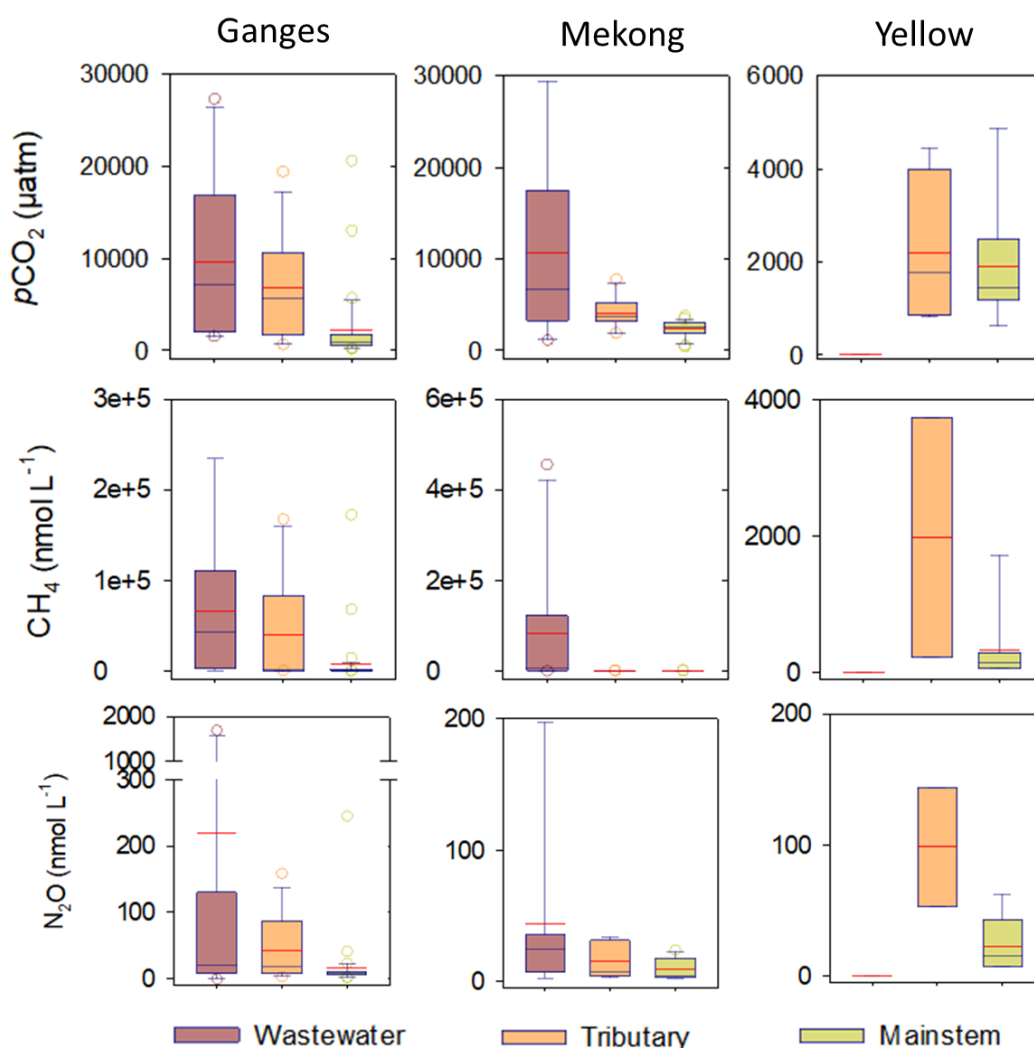


Figure 6. Variations in dissolved GHGs in the Ganges, the Mekong, and the Yellow River. Gas samples were collected from wastewater (n = 22, Ganges: 9–10, Mekong: 7–12, Yellow: 0), tributary (n = 30, Ganges: 13–14, Mekong: 9–12, Yellow: 2–4) and mainstem (n = 60, Ganges: 34–35, Mekong: 12–17, Yellow: 7–9) locations of each river.

The large spatial variations in dissolved GHGs and OM in the three rivers stem from exceptionally high values found in wastewater drains and urban tributaries (Figure 6). Dissolved GHGs were found highest in the wastewater drains, which are discharged to urban tributaries elevating the tributary GHG levels. Distinguished patterns found in the three river systems suggest that the urban tributaries carrying untreated wastewater may significantly contribute to downstream increases in dissolved GHGs, especially in large rivers traversing large metropolitan areas. Levels of GHGs were lower in the Yellow River compared to the Ganges and the Mekong, probably due to better management of wastewater in China. In the Yellow River basin, agricultural irrigation and erosion from arid areas such as Loess Plateau have been suggested as dominant factors determining riverine fluxes of organic C and GHGs (Ran et al., 2015; Park et al. 2018).

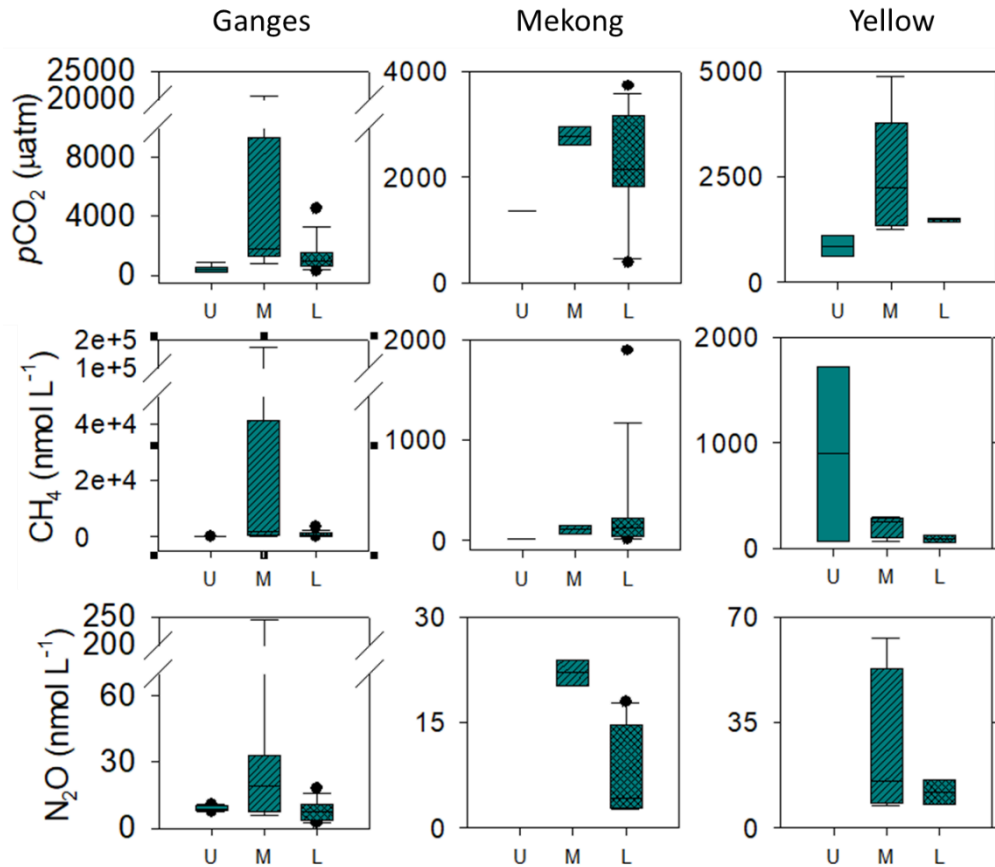


Figure 7. Longitudinal variations in dissolved CO₂, CH₄ and N₂O among the upper (U), middle (M), and lower (L) reaches of the mainstem Ganges (n = 34–35), Mekong (n = 12–17), and Yellow River (n = 7–9).

Dissolved GHGs tended to increase downstream along the middle and lower reaches of the three rivers when the mainstem values were compared longitudinally (Figure 7). This longitudinal pattern may reflect the combined effects of increasing levels of anthropogenic activities such as agriculture, urban wastewater discharge and impoundments along the middle and lower reach on balance between the production and consumption of dissolved GHGs. The only exception from this trend was the highest concentrations of CH₄ measured in the upper reach of the Yellow River, where large expanses of wetlands might function as sources of CH₄ to local streams feeding into the upper Yellow River. The wide ranges of all three GHGs in the middle reach of the Ganges may be attributed to the dominant effect of untreated wastewater discharged from large cities such as Delhi, Agra, Kanpur, Allahabad and Varanasi in the most densely populated river basin in the world.

Van Krevelen diagrams summarizing molecular formulas detected by FT-ICR-MS also exhibited clear longitudinal variations in the molecular-level composition of DOM in the three studied rivers (Figure 8). The upstream samples showed a much smaller number of organic molecules compared to the middle- and lower-reach samples in the Ganges and the Mekong. The middle- and lower-reach samples in all three rivers showed similar organic molecular composition with urban tributaries which contain large number of N and S containing molecules common to anthropogenic organic matter such as fertilizer, impounded water, wastewater and surfactants and biodegraded metabolites (Figure 8). Similar to the levels of dissolved GHGs, the number of organic molecules in the van Krevelen diagram was relatively large in the upper reach of the Yellow River compared to those measured for the upper reaches of the other rivers. The dominance of S containing molecules, which is common to wetland DOM samples (Hertkorn et al., 2016), is also consistent with the high concentration of CH₄ reflecting the influence of wetlands (Figure 7).

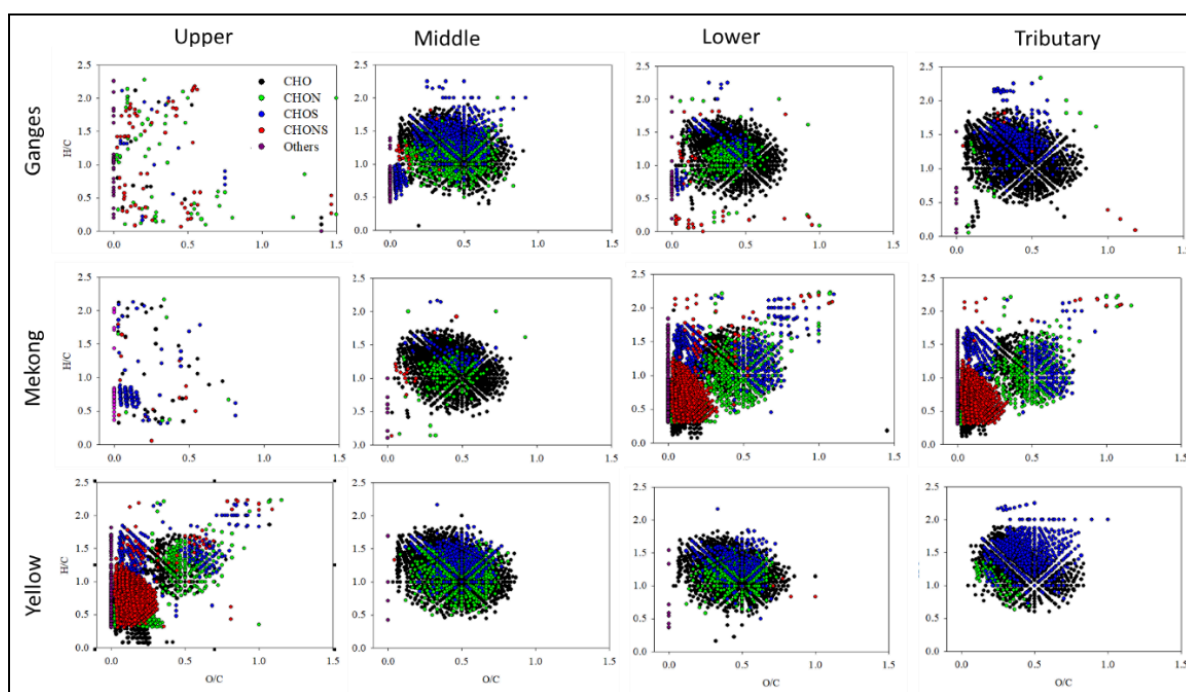


Figure 8. Van Krevelen diagrams showing molecular composition of dissolved organic matter collected in the upper, middle, and lower reaches of the three Asian rivers.

Optical indices such as HIX, FI, and BIX exhibited some longitudinal patterns and clear differences in DOM optical properties among the mainstem, tributary and wastewater samples (Figure 9). These indices together with the relative fraction of three FDOM components identified by PARAFAC, i.e., humic-like (C1), microbial humic-like (C2) and protein like (C3), exhibited clear differences in DOM optical properties among the mainstem, tributary and wastewater samples (Figure 10). Downstream along the upper to middle reaches of the Ganges mainstem, HIX indicative of terrestrially derived DOM tended to decrease, but indicators of autochthonous and/or anthropogenic DOM (FI, BIX and C3) generally increased. However, opposite or no clear trend was observed along the middle to lower reaches of the Ganges and across the Mekong and the Yellow River (Figure 9). Limited sampling coverage in the Mekong and Yellow River might have contributed to the observed unclear longitudinal trends contrasting with the reversing trends of terrestrial vs autochthonous DOM found in other river systems around the world (Massicotte et al., 2017).

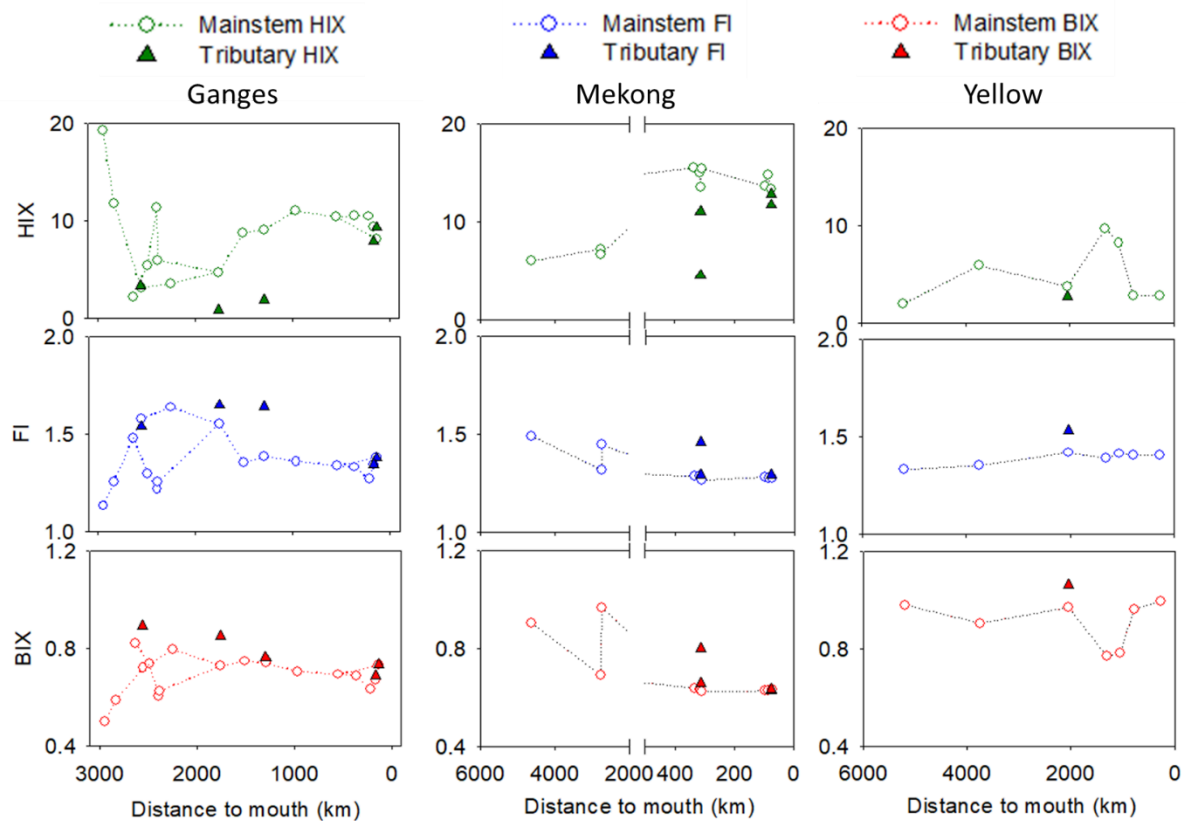


Figure 9. Longitudinal variations in key optical properties of DOM (HIX: humification index; FI: fluorescence index; BIX: biological index) in the Ganges (left), Mekong (middle), and Yellow (right).

Principal component analysis with the optical properties of DOM and dissolved GHGs showed > 50% of the variation explained by the first two principal components (Figure 10). Dissolved GHGs showed positive correlations with DOC, FI and dissolved PO_4^{3-} , consistent with previous studies that observed concomitant increases in labile DOM components and dissolved GHGs (Jin et al., 2018; Begum et al., 2019). Many wastewater samples on the PCA bi-plot were separated from the main stem and tributary samples that overlapped substantially (Figure 10).

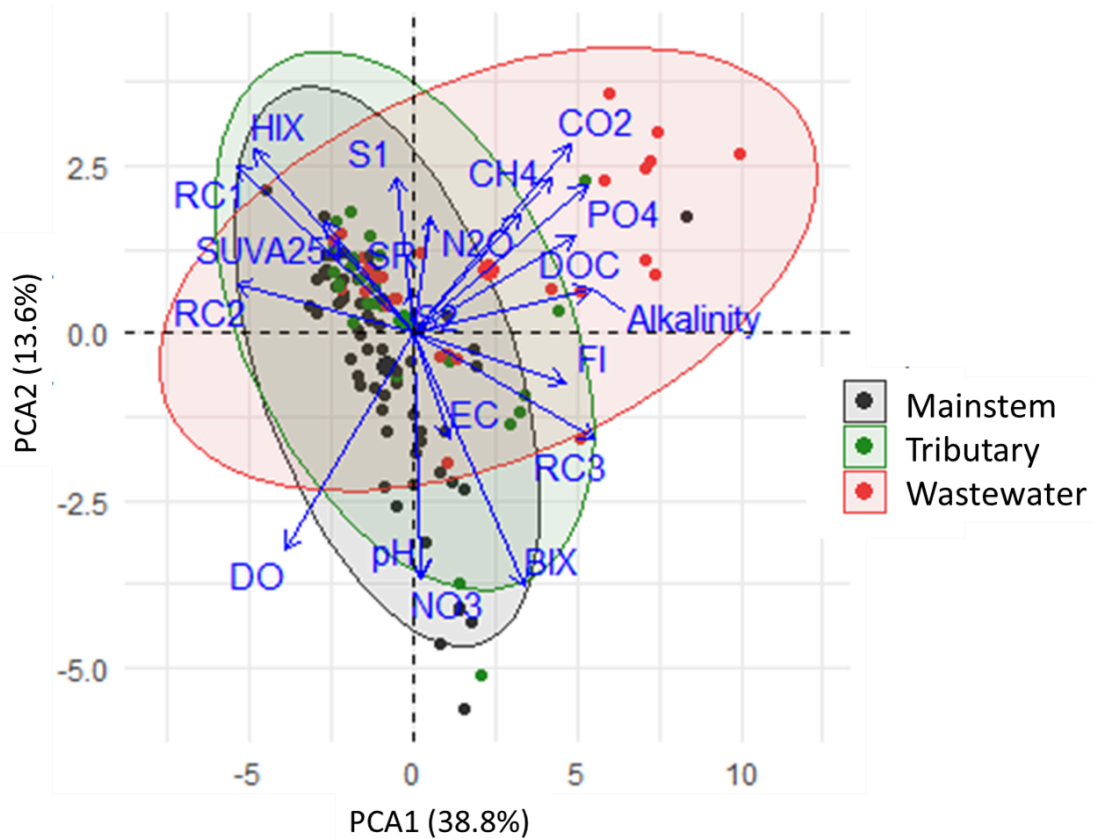


Figure 10. Principle component analysis with dissolved GHGs, DOM and water quality measurements in the mainstem, tributary and wastewater in the Ganges, the Mekong and Yellow River.

3.3.2. Exploratory field studies

(1) Fieldworks in Phnom Penh

- **Cruise underway measurements of $p\text{CO}_2$ and water quality**

Cruise underway measurements of $p\text{CO}_2$ and basic water quality parameters conducted along the Mekong-Tonle Sap River near the confluence revealed the effects of urban wastewater discharged from the metropolitan Phnom Penh on the water quality and the levels of GHGs along the affected river reaches. Urban wastewater drains and downstream rivers transport inorganic nutrients and organic C of anthropogenic origin, contributing to increasing levels $p\text{CO}_2$ (Figure 11). This anthropogenic alteration of riverine C was evident in the underway cruise measurement in the lower reach of Mekong River with a distinct increase in dissolved CO_2 downstream of the confluence with Tonle Sap River (Figure 11).

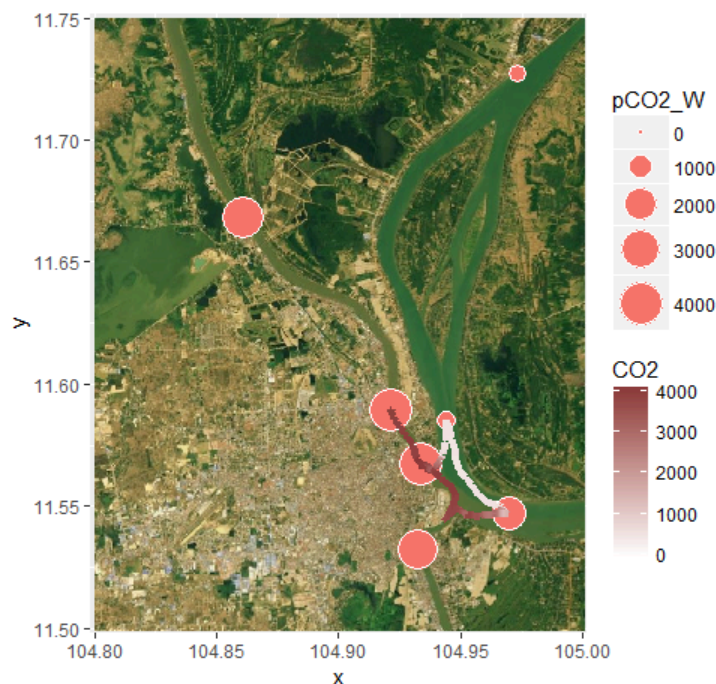


Figure 11. Underway measurements of $p\text{CO}_2$ along the Tonle Sap-Mekong in Phnom Penh, Cambodia. Light- and dark-coloured transects represent underway measurements of $p\text{CO}_2$ using an equilibrator along the Mekong and Tonle Sap, respectively, circles show spot measurements at three pollution sources and four mainstem locations.

- **Field incubation experiment**

A 3-day in situ incubation with river waters from the mainstem and river water mixed with a wastewater sample showed altered patterns of BDOC and dissolved CO_2 in the mixture sample (Figure 12). Continuous measurements of $p\text{CO}_2$ in the headspace of the incubation bottles showed higher rates of CO_2 increase in the headspace in the descending order of the mixture, downstream, and upstream samples under both dark and light conditions (Figure 12). Comparison of nocturnal CO_2 increases between the light and dark incubation samples suggest that inputs of urban sewage in the downstream and mixture samples accelerate the rate of CO_2 increase particularly under light conditions, presumably as a consequence of potential priming effect of newly fixed carbon compounds by phytoplankton (Bianchi et al., 2015; Begum et al., 2019).

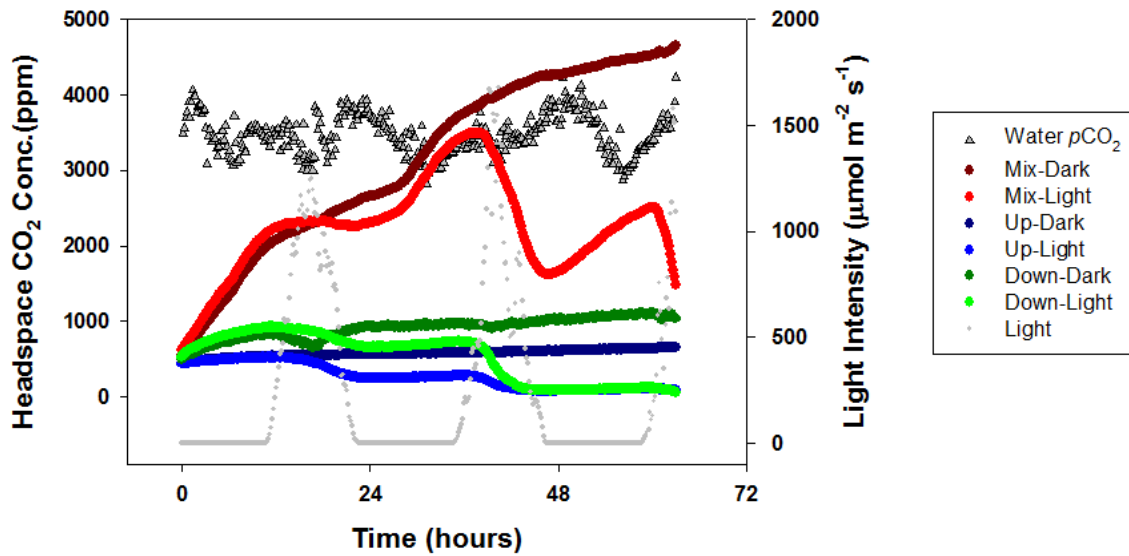


Figure 12. Changes in headspace $p\text{CO}_2$ during a 3-day incubation of the water samples from two locations (“Up” and “Down” sites of the Mekong-Tonle Sap confluence) of the Mekong River and a 10% mixture of an urban wastewater drain (with Up) with and without light exposure.

(2) Fieldworks in Dhaka

• Cruise underway measurements of $p\text{CO}_2$ and water quality

Cruise underway measurements of $p\text{CO}_2$ and basic water quality parameters conducted along the Buriganga-Meghna-Ganges revealed noticeable effects of urban wastewater discharged from the metropolitan Dhaka on the water quality and CO_2 levels along the affected downstream reaches of the tributaries and mainstem Ganges (Figure 13). Urban wastewater drains and tributaries transport loads of inorganic nutrients and organic C of anthropogenic origin, elevating the level of $p\text{CO}_2$ in downstream rivers substantially despite the dilution by larger volumes of river water with lower concentrations of CO_2 . This exploratory study in the lower Ganges illustrated clear contributions of largely untreated urban wastewater from local pollution sources to downstream increases in $p\text{CO}_2$ along the Meghna River near the confluence with Buriganga (Figure 13). The observed levels of $p\text{CO}_2$ in the Meghna and lower Ganges were much higher than some reported values that had been calculated from secondary water quality data (Manaka et al., 2015), suggesting that GHG emissions from the lower Ganges might have been underestimated due to the lack of field measurements.

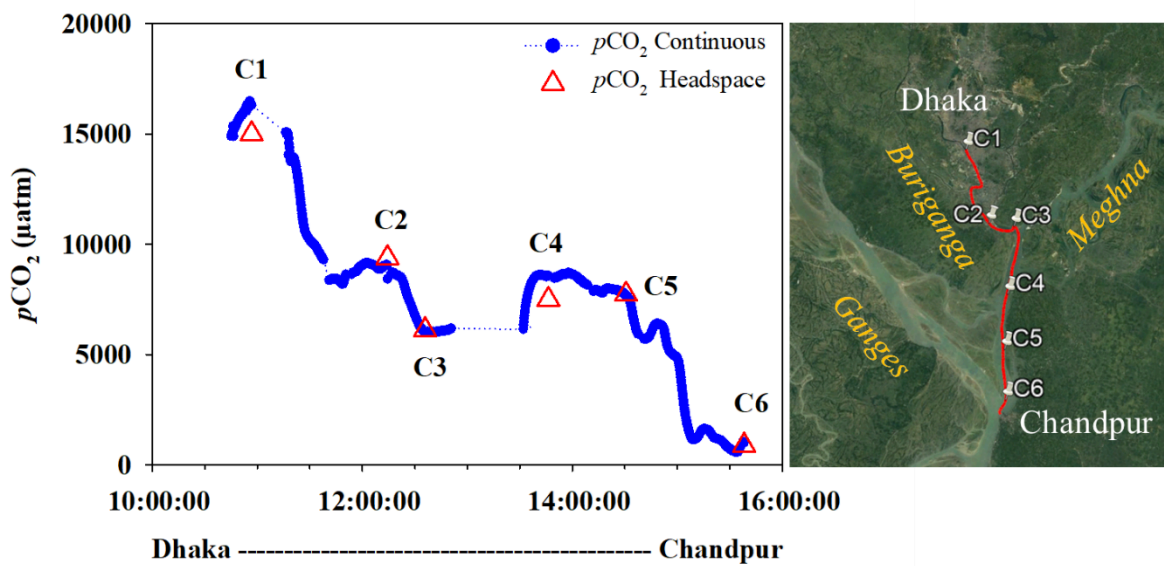


Figure 13. Continuous underway measurements of $p\text{CO}_2$ along the Buriganga-Meghna-Ganges near Dhaka, Bangladesh during a cruise expedition in February 2018.

- **Field incubation experiment**

A 3-day in situ incubation experiment in the lower Ganges, which included a river water and a wastewater sample in isolation or mixed in 9 to 1 ratio (river water: wastewater), showed large differences in BDOC and dissolved CO_2 among the treatments (Figure 14). BDOC measurements showed higher values in the mixture compared to the mainstem samples under both light and dark conditions. The mainstem samples showed negative BDOC concentrations during the incubation under light conditions, indicating increases in DOC concentrations, presumably due to new DOM inputs from phytoplankton biomass. However, the addition of wastewater increased the levels of BDOC and $p\text{CO}_2$ above the values estimated from a conservative mixing of wastewater and river water. This result is consistent with the in-situ incubation experiment conducted in the Mekong River near Phnom Penh (Figure 12) and other studies (Bianchi et al., 2015; Begum et al., 2019), corroborating wastewater-induced priming effect on OM biodegradation in the river receiving loads of labile DOM derived from anthropogenic sources.

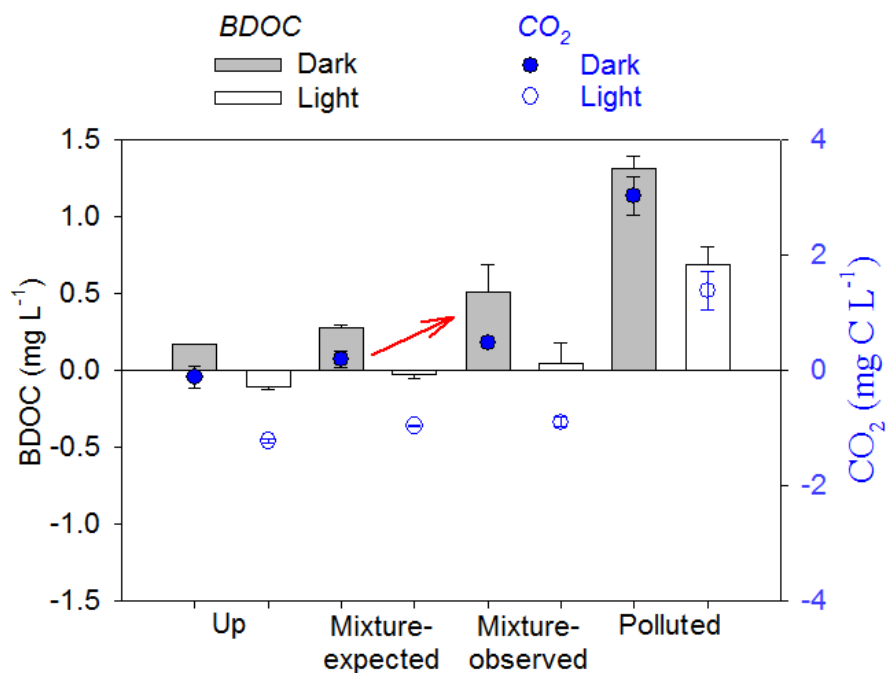


Figure 14. BDOC measured during a 3-day in situ incubation with river water and wastewater samples from the lower Ganges River basin near Dhaka, Bangladesh. Incubation was conducted with or without light exposure.

The molecular composition of DOM in the incubated samples showed complex changes in terms of production and consumption of DOM constituents under light and dark conditions (Figure 15). Active microbial degradation of DOM, as indicated by decreases in peak intensities of detected molecules, resulted in the consumption dominating over the production of DOM constituents in the mainstem and mixture samples under the dark condition. In contrast, the combination of bio- and photodegradation coupled with autochthonous production resulted in a complex array of molecular transformations under the light condition: the production of new molecules dominating in the mainstem and wastewater samples, but consumed molecules exceeding produced molecules in the mixture samples. The mixture sample showed degradation of larger number of organic molecules compared to the mainstem samples under both dark and light conditions. Furthermore, the rate of net production of organic molecules during the incubation was lower in the mixture samples compared to the main samples under the light condition, presumably due to enhanced biodegradation of the wastewater-derived DOM constituents of newly produced organic molecules from photodegradation. Even though organic molecules produced from photodegradation are considered to be refractory (Sankar et al., 2019; Obernosterer et al., 1999), the addition of anthropogenic OM from urban pollution sources might have enhanced biodegradation resulting in the lowered net production, indicating a priming effect of wastewater on the biodegradation of DOM. This result is consistent with BDOC results in Figure 14 and a previous study that reported wastewater-enhanced DOM biodegradation (Begum et al., 2019).

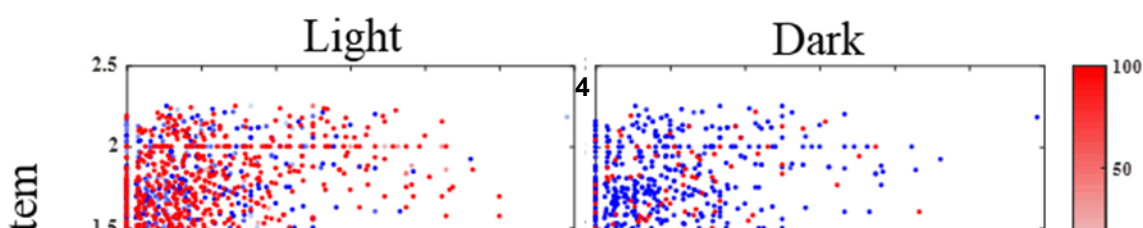


Figure 15. Van Krevelene diagrams showing production and consumption of organic matter during a 3-day in situ incubation with river water and wastewater samples from the lower Ganges River basin near Dhaka, Bangladesh. Incubation was conducted with light (left) or without light (right).

- **One-year in-situ continuous monitoring of $p\text{CO}_2$ in the lower Ganges**

During the 1-year monitoring from May 2018 to April 2019, $p\text{CO}_2$ in the lower Ganges near Kushtia, Bangladesh showed large seasonal variations with highest values recorded during the monsoon season from August-September 2018 (Figure 16). This 1-year monitoring involving continuous in situ sensor measures of $p\text{CO}_2$ for 12 to 48 hours during each sensor deployment represents the first-ever, semi-continuous measurements of riverine CO_2 in the Ganges. Water quality parameters such as pH and DO showed reversed seasonal patterns, with lowest values in monsoon and highest values during pre-monsoon season. Lowered values of DO coinciding with increases in $p\text{CO}_2$ during high flow periods suggest that enhanced biodegradation of riverine OM consuming DO, together with soil-derived $p\text{CO}_2$ in floodwater, might explain the large monsoonal increase in $p\text{CO}_2$, as observed in Indian estuaries (Sarma et al., 2011, 2012). Diel pattern of $p\text{CO}_2$ was also observed during 48-hour continuous monitoring in March 2019 with values ranging from 500-700 ppm (data not shown).

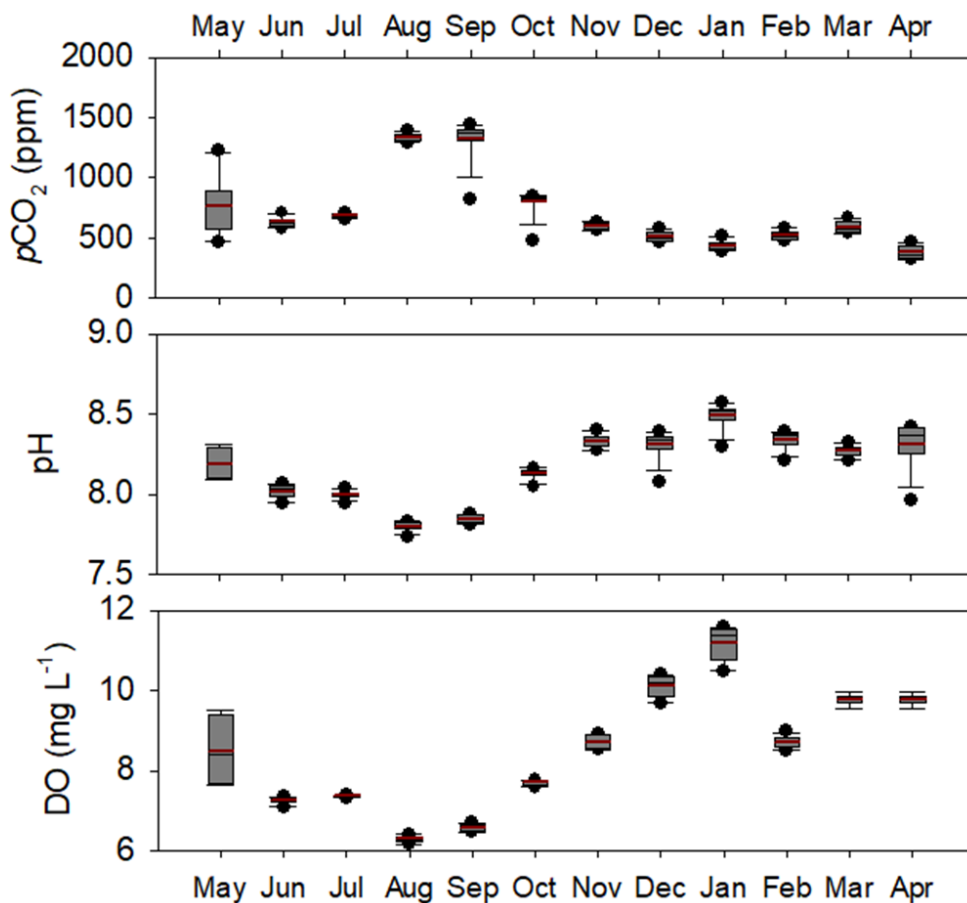


Figure 16. Seasonal variation in $p\text{CO}_2$, pH and DO in the lower Ganges near Kushtia, Bangladesh over the period from May 2018 to April 2019.

3.4. Data compilation toward a regional synthesis

3.4.1. Comparison of $p\text{CO}_2$ measurements and calculations using CO2SYS

Field measurements of $p\text{CO}_2$ in the Ganges, the Mekong and the Yellow River were compared with the values calculated from pH and alkalinity using an aquatic C equilibrium model called CO2SYS to improve the capability of prediction for regional synthesis of riverine $p\text{CO}_2$ based on secondary water quality data available from local sources. The calculation of riverine $p\text{CO}_2$ from temperature, pH and total alkalinity has widely been used in estimating riverine CO_2 outgassing due to the lack of directly measured data (Raymond et al., 2013; Lauerwald et al., 2015). However, recent studies have raised the possibility that these estimated values might be overestimated or underestimated depending on chemical characteristics of river waters such as high organic acidity (Abril et al., 2015).

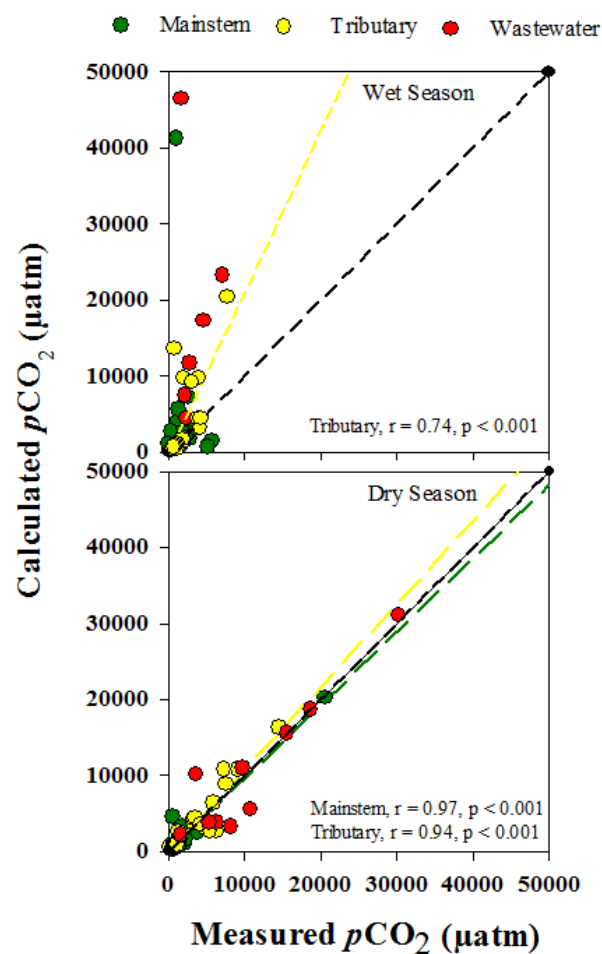


Figure 17. Correlation between measured (headspace equilibration method) and calculated (CO2SYS) values of riverine $p\text{CO}_2$ (μatm), analysed separately for wet and dry seasons. Samples were collected from the mainstem (n: 33 for wet season; 20 for dry season), tributaries (n: 29 for wet season; 30 for dry season) and wastewater (n: 7 for wet season; 9 for dry season) of three Asian rivers (the Ganges, Mekong and Yellow River) from 2015 to 2019. Only significant correlation ($P < 0.05$) is indicated by a dotted regression line through the plot.

Calculated and measured $p\text{CO}_2$ values exhibited relatively good or bad agreements depending on the season (Figures. 17, 18). The spatial and seasonal patterns of calculated vs measured $p\text{CO}_2$ along the three rivers exhibited good agreements during the dry season and bad agreements during the wet season for three different water types (mainstem, tributary and wastewater). Figure 16 illustrates strong positive correlations between the values of $p\text{CO}_2$ measured by headspace equilibration method and calculated by CO2SYS program during the dry season, for all water types ($r > 0.90$; $P < 0.001$) except wastewater ($r = 0.54$), although during the wet season significant correlation was found only for the tributary samples ($r > 0.70$; $P < 0.001$). Low pH values during the wet season might have created conditions responsible for large discrepancies between calculated and measured values of $p\text{CO}_2$ in the monsoonal rivers characterized by strong seasonality in precipitation and river flow.

Abril et al. (2015) ascribed an overestimation of $p\text{CO}_2$ in acidic, organic-rich inland waters by 50 to 300% to the combined effect of organic acids contribution to alkalinity and the limited buffering capacity of the carbonate system. Potential overestimation of $p\text{CO}_2$ in floodwater containing high levels of organic C during the monsoon period implies that some caution and corrective measures should be taken when employing C equilibrium models in estimating $p\text{CO}_2$ across various sites, particularly during high flow periods. Based on this result, we recommend a constrained use of C equilibrium models when estimating riverine $p\text{CO}_2$ during the wet season. It remains unresolved whether corrective measures such as adjusting pH to control bias associated with low pH values could reduce uncertainty levels of pH-based $p\text{CO}_2$ calculations.

When CO2SYS calculations were compared with $p\text{CO}_2$ measurements obtained using three methods used for underway measurements during a cruise expedition along the Buriganga-Meghna-Ganges in February 2018, all three measurements exhibited strong positive correlations with the calculated values (Figure 18): equilibrator ($r = 0.99$; $P < 0.001$), PTFE membrane-enclosed CO_2 sensors ($r = 0.98$; $P < 0.001$) and headspace equilibration ($r = 0.99$; $P < 0.001$). These results confirm the accuracy of the three employed $p\text{CO}_2$ measurement techniques, consistent with the previous tests in a different river system in Korea (Yoon et al., 2016). The strong correlations between the measured and calculated values of $p\text{CO}_2$ in the lower Ganges and tributaries, which were found in a dry season (February 2019), also corroborate the significant relationships between the calculated and observed values obtained from the three Asian rivers during the dry season (Figure 17).

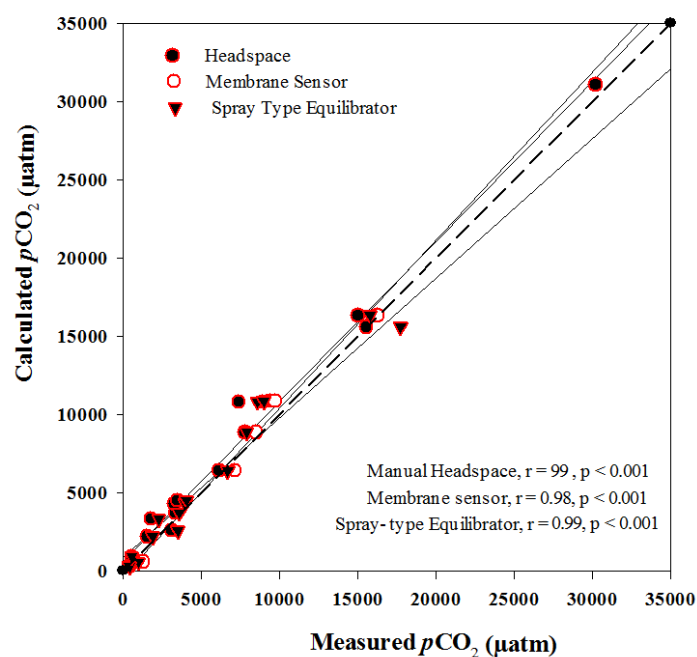


Figure 18. Correlations between measurements of riverine $p\text{CO}_2$ (μatm) using three different methods (PTFE membrane–enclosed CO_2 sensor, spray-type equilibrator and manual headspace equilibration) and calculated values using CO2SYS. Field measurements were obtained from 16 sampling locations along the main stem and tributaries of the lower Ganges near Dhaka, Bangladesh during a cruise expedition in February 2018 and Mekong and Tonle Sap along with polluted tributaries Phnom Penh, Cambodia during a cruise expedition in 2017.

3.4.2 Estimating riverine organic C and GHGs from BOD data

Riverine organic pollution results from the land-derived input of easily biodegradable organic materials into surface waters via the discharge of domestic, industrial and agricultural runoff from various anthropogenic sources. Increasing loads of untreated wastewater can alter the rate of OM biodegradation, often increasing emissions of GHGs (CO_2 , CH_4 and N_2O) from water to the atmosphere. Biochemical oxygen demand (BOD) can be used as an indicator of organic pollution and further to estimate riverine concentrations of DOC, TOC and GHGs (CO_2 , CH_4 and N_2O). Given high data availability of BOD as a common water quality parameter included in many national monitoring programs, BOD-based estimates of DOC, TOC and GHGs can contribute to reducing uncertainties in estimating regional-scale riverine C fluxes and GHG emissions from Asian rivers.

To calculate BOD-based TOC, we assumed a conversion factor of 0.7 for BOD/TOC in the Ganges mainstem from headwaters to the lowest reaches and a conversion factor of 1.94 for BOD/TOC in wastewater drains and highly polluted tributaries, based on ratios found in the literature (Table 6). A conversion factor of 0.85 and 2.7 for BOD/DOC was used to calculate BOD-based DOC for the Ganges mainstem and wastewater drains or polluted urban tributaries, respectively. A conversion factor of 0.9 and 1.6 for DOC/ CO_2 was established from field measurements in the Ganges basin during the years 2017–2018 (Figure 19) to calculate BOD-based CO_2 for the Ganges mainstem and wastewater drains or polluted urban tributaries, respectively.

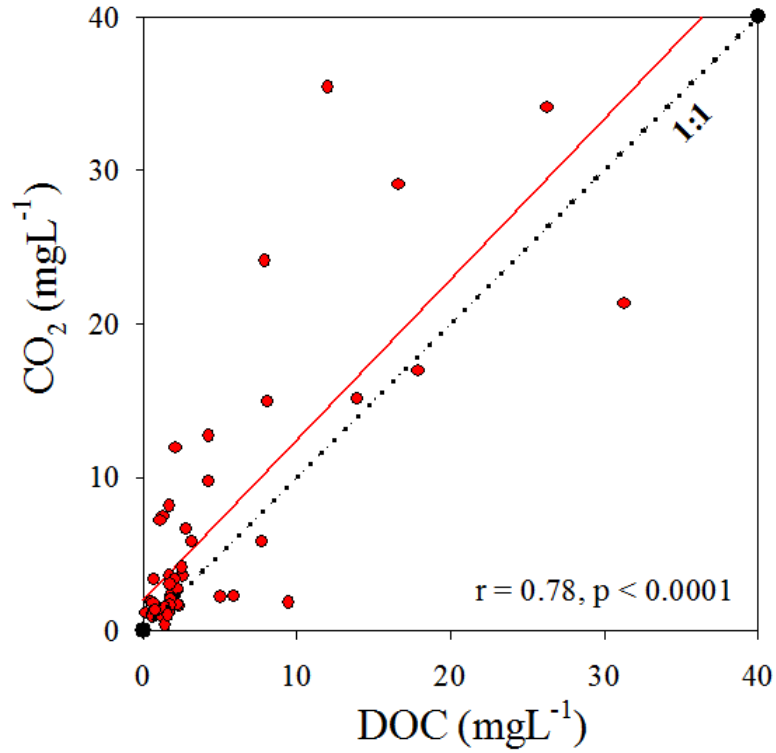


Figure 19. Correlation between CO₂ and DOC concentrations measured in the Ganges River in 2017–2018.

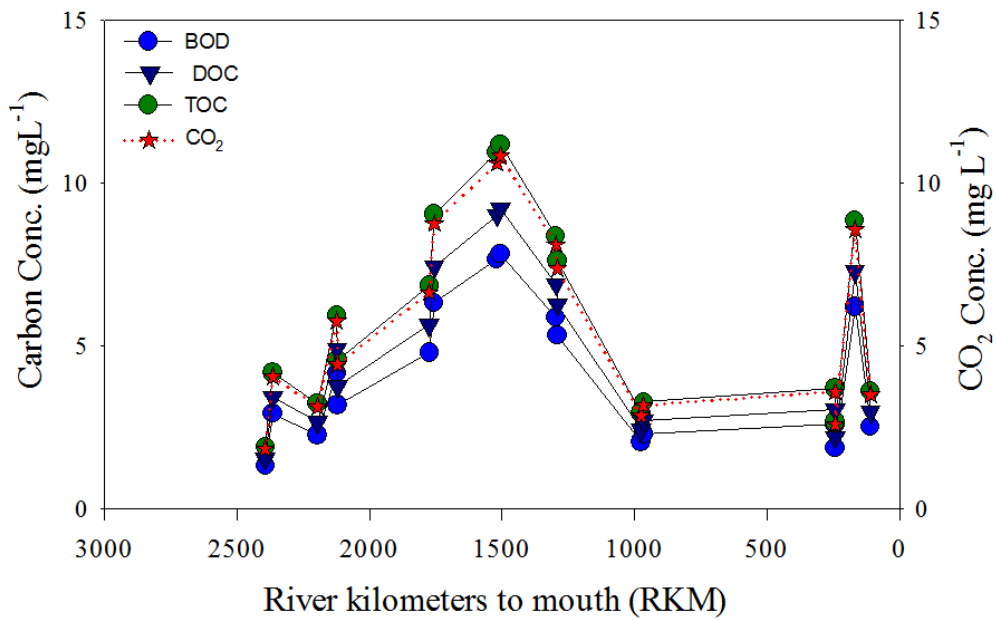


Figure 20. Longitudinal variations in average BOD, DOC, TOC and CO₂ concentrations in 1986–2014 along the Ganges mainstem from the upper to most downstream reaches.

Table 6. BOD₅/TOC and BOD₅/DOC river ratios from literature.

Author	BOD ₅ /TOC	BOD ₅ /DOC	Location	Notes
Lee et al. 2016	0.64		4 watersheds in Rep. of Korea	Ratio of mean values of 114 river stations in 4 watersheds
Ebise and Inoue 1991.	0.85, 0.72		River Hinuma, inflow to Lake Kasumigaura, Japan	Mean and discharge-weighted, respectively
Jung et al. 2016	0.51		Nakdong River watershed, Rep. of Korea	Mean of mean values from 20 monitoring stations
Viera et al. 2012	1.29		Lis River and tributaries, Portugal	Mean of several river stations
Ouyang, 2005	0.12		St Johns River, Florida, USA	Mean of all stations
Benfenati, E. et al. 1992	0.91		Adige R Italy	Mean of 2 river campaigns
Pradhan et al. 2015	0.23	0.29	Hindon River, a tributary of Yamuna River, India	Mean of several river stations
Serajuddin et al. 2018	1.22	2.9	Sitalakhya River, a tributary of Ganges River, Bangladesh	Mean of river station in different time, January to April 2011
Haritash et al. 2016	0.4		Rishikesh, upstream of Ganges River, India	Mean of 19 stations along the Rishikesh known as upper reach of Ganges river
Ministry of Environment, Republic of Korea	0.73		Han River, South Korea	Mean of 13 sites of Han River (2008–2018)
Leithold et al. 2017		1.55	Iguacu River, Brazil	Mean of several river station without urban influences
Leithold et al. 2017		4.53	Iguacu River, Brazil	Mean of several river station with urban influences
Lim, 2019		0.85	Han River and tributaries, South Korea	Mean of 18 sites in the Han River and tributaries
Quayle et al. 2009	2.3	2.7	Winery wastewater	

Table 7. BOD-based estimates of TOC, DOC and CO₂ concentrations for the mainstem sites of the Ganges. The distance from the river mouth is expressed in river km (RKM). Different measurement periods are indicated by the years in the heading.

Site (RKM)	Concentration (mg L ⁻¹) for 1986				Concentration (mg L ⁻¹) for 2002				Concentration (mg L ⁻¹) for 2012-2014			
	BOD	TOC	DOC	CO ₂	BOD	TOC	DOC	CO ₂	BOD	TOC	DOC	CO ₂
2394	1.70	2.43	2.00	2.35	1.71	1.41	1.66	1.71	1.10	1.57	1.29	1.52
2365	1.80	2.57	2.12	2.49	2.43	2.00	2.35	2.43	5.30	7.57	6.24	7.34
2197	2.20	3.14	2.59	3.04	3.00	2.47	2.91	3.00	2.50	3.57	2.94	3.46
2120	5.50	7.86	6.47	7.61	1.71	1.41	1.66	1.71	2.90	4.14	3.41	4.01
2123	5.10	7.29	6.00	7.06	6.00	4.94	5.81	6.00	3.20	4.57	3.76	4.43
1775	7.20	10.29	8.47	9.97	5.43	4.47	5.26	5.43	3.40	4.86	4.00	4.71
1757	8.60	12.29	10.12	11.90	7.00	5.76	6.78	7.00	5.50	7.86	6.47	7.61
1520	11.40	16.29	13.41	15.78	11.43	9.41	11.07	11.43	3.60	5.14	4.24	4.98
1506	15.50	22.14	18.24	21.45	5.43	4.47	5.26	5.43	4.20	6.00	4.94	5.81
1296	2.50	3.57	2.94	3.46	15.14	12.47	14.67	15.14	4.50	6.43	5.29	6.23
1291	3.00	4.29	3.53	4.15	14.43	11.88	13.98	14.43	2.90	4.14	3.41	4.01
976	2.00	2.86	2.35	2.77	2.71	2.24	2.63	2.71	2.30	3.29	2.71	3.18
966	2.20	3.14	2.59	3.04	2.86	2.35	2.77	2.86	2.70	3.86	3.18	3.74
109	2.20	3.14	2.59	3.04	2.86	2.35	2.77	2.86	3.40	4.86	4.00	4.71
242	1.90	2.71	2.24	2.63	1.57	1.29	1.52	1.57	2.63	3.76	3.09	3.64
243	No data	No data	No data	No data	No data	No data	No data	No data	2.60	3.71	3.06	3.60
168	No data	No data	No data	No data	No data	No data	No data	No data	6.20	8.86	7.29	8.58

BOD data collected across the Ganges over three decades (1986–2014; data source: CPCB and other scientific publications) showed distinct longitudinal variations along the upper, middle and lower reaches (Table 7). BOD-based estimates of TOC, DOC and CO₂ along the Ganges mainstem revealed lower levels of TOC, DOC and CO₂ in the upper reaches of the river compared to the middle to lower reaches (Figure 20; Table 7). Very high levels of OM and CO₂ estimated for the middle reach reflected the effect of poorly treated industrial and domestic wastewater discharged from large metropolitan cities and industries. Wastewater drains and canals and highly polluted urban tributaries export loads of labile OM of anthropogenic origin to the middle and lower reaches of the Ganges (Table 8). Annual loads of BOD, DOC and TOC feeding into the Ganges mainstem in India amount to 364.6×10⁹ g, 135.1×10⁹ g, and 188.0×10⁹ g, respectively (Table 8). High loads of labile organic matter of anthropogenic origin can alter the rate of biodegradation of riverine organic matter in downstream rivers, also enhancing the rate of CO₂ emission from water to atmosphere, as observed in the Hooghly estuary (Samanta et al., 2015) and other Indian estuaries (Sarma et al., 2012).

Table 8. Annual loads of BOD and BOD-based DOC and TOC calculated for the upper, middle and lower reaches of the Ganges basin. Two lower reaches refer to the Hooghly in India and the Padma-Meghna in Bangladesh.

Country	River reaches	No. of Drains	Flow (MLD) ×10 ²	BOD load (g yr ⁻¹) ×10 ⁹	TOC load (g C yr ⁻¹) ×10 ⁹	DOC load (g C yr ⁻¹) ×10 ⁹
India	Upper	14	4.4	15.3	7.9	5.7
	Middle	70	38.7	313.9	161.8	116.3
	Lower	54	17.7	35.4	18.3	13.1
Total loads				364.6	188.0	135.1

3.4.2. Combining literature data with field measurements to estimate riverine CO₂ emissions from the Indian subcontinent

The study sites included major rivers of the Indian subcontinent, namely the Ganges, Brahmaputra, Indus, Krishna, Godavari, Narmada, Mahanadi, Tapti, Bhote Koshi and other various inland waters of India (Figure 21; Table 9). Data were obtained not only for mainstem locations but also for headwaters and tributaries feeding into the mainstem.

Table 9. Summary of field samplings conducted in the 9 major rivers in the Indian subcontinent.

River	Component	Country	Distance to mouth (km)	Sampling points	Sampling time
Brahmaputra	Headwater	China		6	2007–2018
	Mainstem	China, Nepal, India, Bangladesh	3277–281	42	
	Tributary	China, Nepal, India, Bangladesh		60	
Ganges	Headwater			6	1982–2018
	Mainstem	India Bangladesh	2763–546 546–168	10 10	
Yamuna	Mainstem	India	2946–1755	8	
Hooghly	Mainstem	India	546–27	35	
Local tributaries	Tributary	Bangladesh		45	
	Wastewater	India Bangladesh		4 4	
Godavari	Headwater	India		1	2009–2012
	Mainstem		1460–258	31	
	Tributaries			11	
	Impoundment in Mainstem		1384–39	6	
Indus Sutlej	Mainstem	India, Pakistan	2456–154	23	1994–1995
	Mainstem		1747–1107	6	
	Tributaries			31	
Krishna	Headwater	India		3	2001–2002
	Mainstem		2536–976	10	
Bhima	Mainstem		1073–731		
	Tributaries			14	
Mahanadi	Mainstem	India	119–240	5	2005
	Tributaries			8	
Narmada	Headwater	India		1	2001–2004
	Mainstem		1266–187	12	
	Tributaries			13	
Tapti	Mainstem	India	443–302	2	2001
Purna	Mainstem		615–430	2	
	Tributaries			7	
Bhote Kosi	Mainstem	India		39	2003

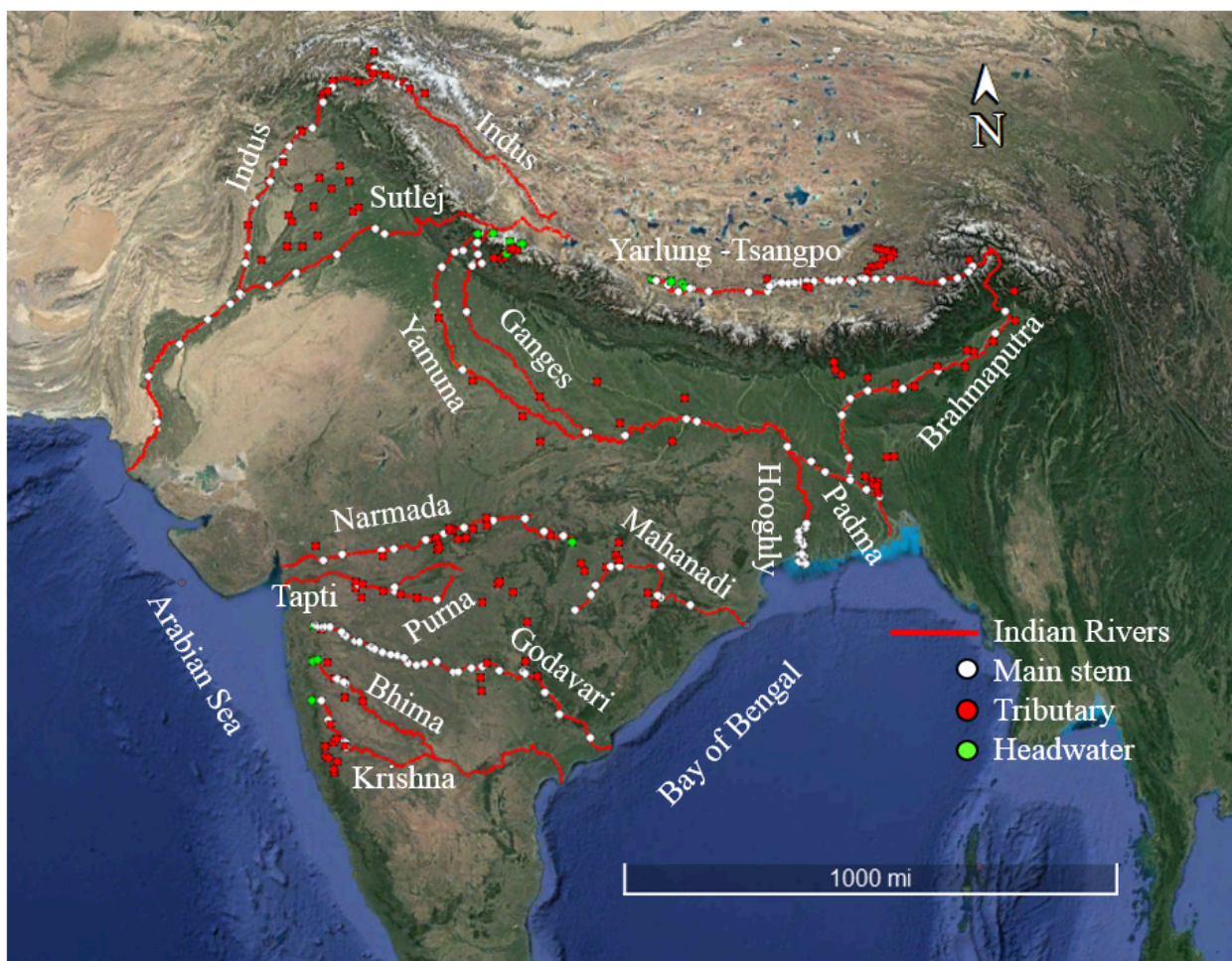


Figure 21. Study sites including the mainstem, headwaters, and tributaries of 8 major rivers in the Indian subcontinent.

- **Geomorphological characteristics and spatial variations in riverine $p\text{CO}_2$ – A synthesis of literature data**

A higher level of $p\text{CO}_2$ was observed along the middle to lower reaches of the mainstems and polluted urban tributaries compared to the upstream reaches both for northern rivers and rivers originating from the Deccan Traps, except for the Godavari (Figure 22). The increasing patterns of $p\text{CO}_2$ along the middle and lower reaches of the Indian subcontinental rivers depicting the input of anthropogenic C from directly pollution sources or polluted urban tributaries feeding the mainstem channel altered the natural riverine C biogeochemistry. Along the Godavari river one of the upper reach site namely 'Shirdi' a famous tourist spot for its religious importance observed as highly polluted site with higher value of $p\text{CO}_2$ ($11581\mu\text{atm}$) due to having large population influx generating extra sewerage load along with agricultural runoff and directly discharged to the river (Jha et al. 2009).

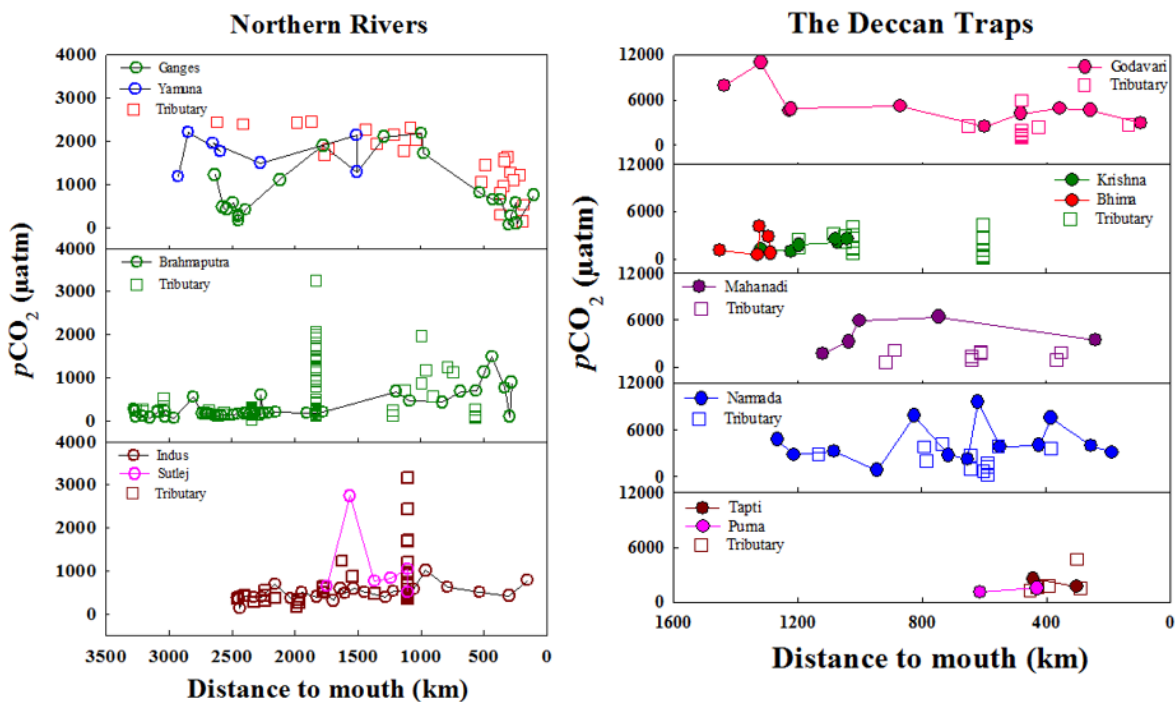


Figure 22. Longitudinal variation in partial pressure of CO₂ ($p\text{CO}_2$) along the river basins in the Indian Subcontinent including northern Rivers (Ganges, Brahmaputra and Indus) and rivers in the Deccan Traps (Godavari, Narmada, Tapi, Krishna and Mahanadi) from headwaters to mouth.

- **Comparison between literature data and field measurements of $p\text{CO}_2$ in the Ganges and Godavari**

Water and gas sampling along the mainstems of the Ganges, Yamuna, Hooghly and Padma in the Ganges basin were performed during July-August 2017, as the wet season, and February 2018, as dry season. Various sampling sites including wastewater drains and polluted urban tributaries draining metropolitan cities represented dominant land uses and watershed characteristics across the Ganges basin, offering opportunities to compare riverine water quality and GHG concentrations between the less populated upper reaches and the more urbanized lower reaches receiving high loads of anthropogenic pollutants. Two seasonal surveys revealed very high $p\text{CO}_2$ value along the Yamuna reach traversing Delhi and Agra and urban tributaries draining large cities across the basin (Figure 23). As compiled in Table 8, numerous wastewater drains across the Ganges basin discharge labile organic materials derived from domestic and industrial wastewater and livestock farming to the Ganges mainstem and urban tributaries, enhancing biodegradation and photodegradation of riverine OM and hence CO₂ emissions along downstream reaches.

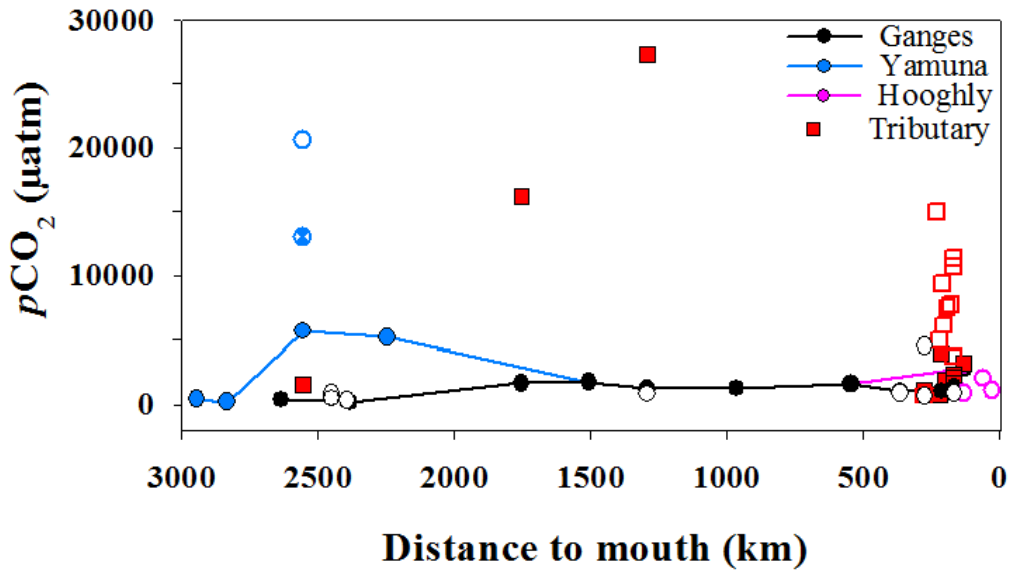


Figure 23. Longitudinal variation in partial pressure of CO₂ ($p\text{CO}_2$) in the Ganges River basin. The data of $p\text{CO}_2$ are from field surveys in August 2017 (Filled symbol), February 2018 (Void symbol) and June 2019 (Semi-filled symbol).

Monsoonal rivers in the Deccan Traps, including the Godavari, Narmada, Tapti, Krishna and Mahanadi, empty into the Bay of Bengal or the Arabian Sea. The Godavari, as one of the largest rivers in India, discharge annually around 70 km³ to the Bay of Bengal. The total length of river is 1600 km and this river has been impounded to meet increasing demands for irrigation and domestic consumption of water. Two seasonal sets of data collected along the Godavari from headwater to mouth exhibited higher $p\text{CO}_2$ during peak discharge periods in monsoon, whereas both discharge and $p\text{CO}_2$ decreased during dry season and impounded reaches also exhibited similar seasonal patterns (Figure 24).

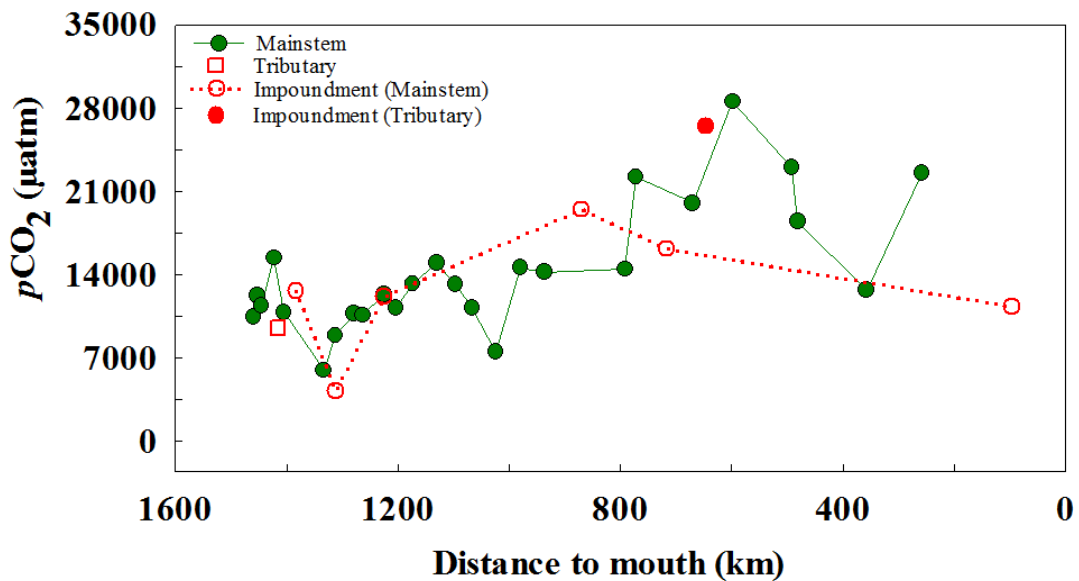


Figure 24. Longitudinal variation in partial pressure of CO₂ ($p\text{CO}_2$) in the Godavari River basin. The data of $p\text{CO}_2$ are from field surveys in 2010–2012.

Large discrepancies in $p\text{CO}_2$ between literature data and recent field measurements across the Ganges and Godavari (Figure 25). Recent field measurements exhibited large spatial variations in $p\text{CO}_2$ across the Ganges and Godavari, including mainstems, tributaries, wastewater drains and impounded reaches (Figure 25), illustrate a longitudinal alteration of riverine organic matter composition corresponding to the level of anthropogenic perturbation. Exceptionally high levels of $p\text{CO}_2$ along the middle and lower reaches of the Ganges (Figure 23) compared to the upper reach and other Indian rivers (Figure 22), along with other recent regional-scale assessments (Park et al., 2018), suggest that more field measurements are required to better assess pollution impacts on riverine emissions of CO_2 and other GHGs from the Ganges river basin and other rapidly urbanizing river basins across the region. Compared with literature data of $p\text{CO}_2$, which usually do not include $p\text{CO}_2$ in wastewater drains and impoundments, the mean $p\text{CO}_2$ from the field measurements across the Ganges and Godavari exhibited 5 times and 3 times higher levels of $p\text{CO}_2$ than the reported values, respectively (Figure 25). This result confirms the previous studies were attributing the potential underestimation of global riverine CO_2 emission to poor spatial coverage and quality of data (Lauwerwald et al., 2015; Park et al., 2018).

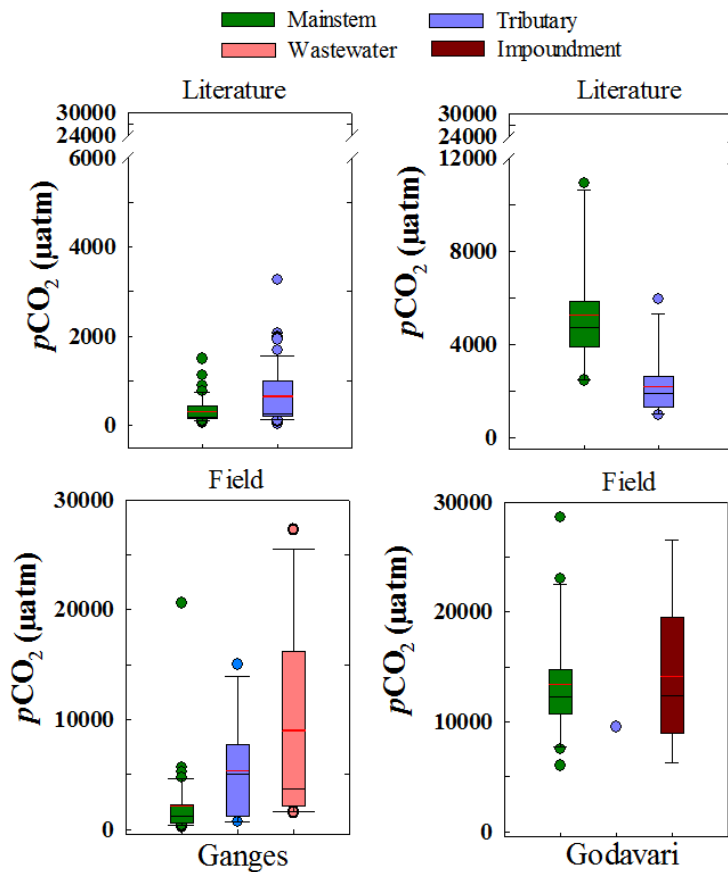


Figure 25. Comparison of literature data and new field measurements of $p\text{CO}_2$ among the mainstem, tributaries, wastewater drains and impounded reaches of the Ganges and Godavari. Ganges field measurements in 2017–2018 and Godavari field measurements in 2010–2012. The horizontal black and red lines of the box plots represent the median and mean values, respectively. Each box covers the 25th to 75th percentile, whereas the whiskers represent the 10th and 90th percentile.

- **Regional-scale synthesis of riverine $p\text{CO}_2$ and CO_2 emissions**

Across the Indian subcontinent, the level of riverine $p\text{CO}_2$ was consistently higher than the atmospheric $p\text{CO}_2$ ($\sim 400 \mu\text{atm}$), with an average of $2521 \mu\text{atm}$ (range: 28 to $28588 \mu\text{atm}$). These compiled values of $p\text{CO}_2$ were combined with estimates of river surface area and gas transfer velocity to produce an average rate of riverine CO_2 emission at $291 \text{ mmol m}^{-2} \text{ day}^{-1}$, with a range of -90 to $3942 \text{ mmol m}^{-2} \text{ day}^{-1}$. The estimated average rate of riverine CO_2 emission exceeds the previously reported value of $163 \text{ mmol m}^{-2} \text{ day}^{-1}$, which was estimated by Li and Bush (2015) for the Indian river basins covering around 52600 km^2 (Table 10). This study provided the average annual riverine C export from the Indian subcontinent at 67 Tg C per year, which exceeds 37 Tg C estimated for the Indian river basins covering around 52600 km^2 (Li and Bush, 2015). During the period from 1980–2000, the annual total C export from South Asian rivers was estimated at 42.9 Tg C , accounting for 7% of the global riverine C export (611 Tg C ; Parta et al., 2013). The average rate of CO_2 emission from the Ganges was estimated at $383 \text{ mmol m}^{-2} \text{ day}^{-1}$ with a range of -26 to $3703 \text{ mmol m}^{-2} \text{ day}^{-1}$ (Table 10). Two seasonal (wet and dry) estimates of CO_2 emissions from the Ganges River exhibited high seasonal variability across the mainstem, tributary and wastewater (Figure 26). The rivers in the Deccan traps of India exhibited higher rates of CO_2 emission compared to other rivers of the Indian subcontinent (Table 10). This study can contribute to reducing the large uncertainty in estimating the global riverine CO_2 emission by providing more spatially resolved data of $p\text{CO}_2$ and CO_2 emissions from major rivers in the Indian subcontinent. In particular, explorative field measurements in the Ganges basin illustrate the importance of more field-based studies to provide a more accurate assessment of pollution impacts on human-modified Asian rivers.

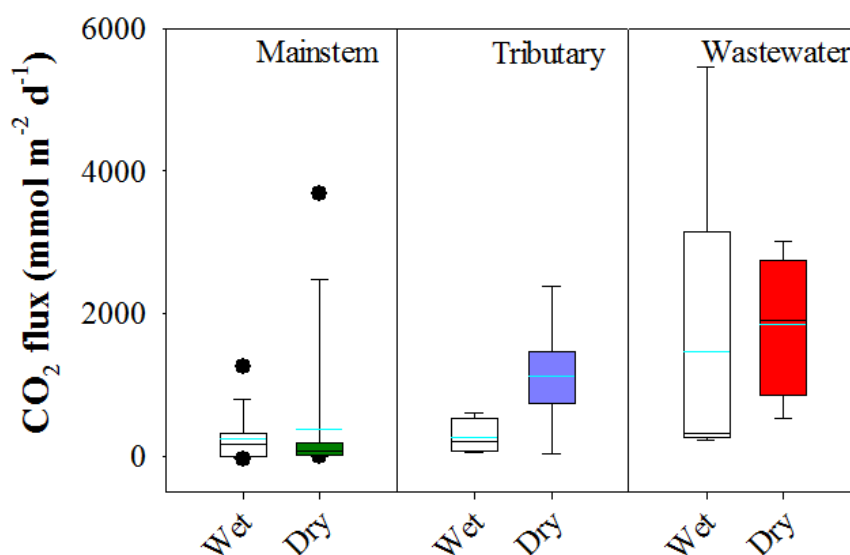


Figure 26. Comparison of new measurements of CO_2 emission among the mainstem, tributary and wastewater of the Ganges River during the wet and dry seasons. The horizontal black and blue-green lines of the box plots represent median and mean, respectively.

Table 10: CO₂ emission with C export from Indian subcontinental rivers and south Asian rivers from various studies of the ongoing decade.

	River	Area km ² × 10 ³	Mean pCO ₂ (range) (µatm)	Mean (range) CO ₂ emission		Tg C yr ⁻¹	References
				k (cm h ⁻¹)	CO ₂ emission (mmol m ⁻² day ⁻¹)		
Indian subcontinent	Brahmaputra		656 (28–6706)	19 (8–27)	18 (–90–520)		This study
	Ganges		2876 (62–20580)	16 (8–24)	383 (–58–3703)		
	Godavari		10152 (969–28588)	12 (11–23)	1132 (79–3403)		
	Indus		694 (134–5907)	11 (5–34)	35 (–35–315)		
	Krishna		1903 (206–4907)	10 (9–10)	155 (–20–405)		
	Mahanadi		4447 (1205–11465)	11	251 (32–726)		
	Narmada		5924 (630–17337)	10 (8–10)	324 (–10–992)		
	Tapti		3447 (2031–8479)	11	179 (86–520)		
	Bhote Kosi		593 (35–5907)	7	15 (–26–441)		
	9 major rivers	52.6			291	67	
Indian subcontinent	52.6			163	37.5	Li and Bush, 2015	
Indian rivers	17.6			20	2.53	Panneer Selvam et al., 2014	

South Asia							

USA							

World	Rivers	357.6			541.1		Butman and Raymond, 2011
	Rivers				146.85		Bastviken et al., 2011; Li et al., 2013
	Rivers & streams	536					Raymond et al., 2013
	Rivers (60–100 m)	220–360					Aufdenkampe et al., 2011
	Streams (60–100 m)	90–150					Aufdenkampe et al., 2011

3.5. Synthesis and science-to-policy translation

3.5.1. Combining literature data with field measurements to assess human impacts on riverine CO₂ emissions from Asian rivers

As literature review (Park et al., 2018) and field surveys in the three river basins illustrated, recent increases in water pollution and ensuing changes in riverine metabolism and GHG emissions cannot be captured adequately by old literature data or individual field studies in a small number of sites covered by conventional routine monitoring. Therefore, we combined the following three approaches to providing more reliable ranges of estimates for organic C and GHG fluxes in major Asian rivers under strong influence of urbanization and water pollution. First, where no direct measurements are available, riverine CO₂ concentrations were calculated using an aquatic C equilibrium model (CO₂SYN) and basic water quality data including water temperature, pH, alkalinity available from a global water quality database (GLORICH: Global River Chemistry Database). In the case of the three studied river basins, calculated values were compared with our field measurements to provide empirical relationships that can improve estimation accuracy. Second, the measured concentrations of the three GHGs were used to update and complement published data on the three rivers. The survey results on the role of tributary pollution as a major GHG source in the lower reaches of the studied rivers were combined with recent reach-scale reports on organic C loads and GHG emissions in separate reviews focusing on pollution and dam impacts on the organic C and GHG fluxes in the Ganges and Mekong.

The last approach benefited from a recent global assessment of surface water quality based on a UNEP-coordinated environmental monitoring program (GEMS; UNEP, 2016). In particular, BOD data available from GEMStat and other local sources and literature were compared with our field measurements of the three GHGs while considering IPCC guidelines for estimating CH₄ and N₂O emissions from wastewater to provide empirical equations for estimating GHG emissions from rivers highly polluted by wastewater. BOD-GHG relationships were compared with our exploratory field study results to evaluate the potentials of stable and labile fractions in organic C loadings as GHG sources.

The combination of these three approaches was applied to the Indian Subcontinent to test and establish frameworks for estimating organic C export and GHG emissions from rapidly urbanizing river basins across Asia. Combining model-based estimation with field measurements of pCO₂ provided a more reliable, regional-scale estimate of riverine CO₂ emission from the Indian Subcontinent, which reflected recent increases in organic loads in wastewater from urbanizing rivers across the region. Ranges of fluxes estimated for large rivers in the Indian Subcontinent can form a base of regional-scale assessment of pollution impacts on organic C export and GHG emissions from all rivers in Asia. Spatially more resolved data obtained from this synthesis will improve the resolution and reliability of the existing global data and hence provide more accurate estimates for anthropogenic C loadings and their contributions to riverine GHG fluxes.

3.5.2. Implications for policies on watershed management and climate change mitigation

A policy-relevant project goal is to provide policymakers and the general public with science-based alternative watershed management options integrating climate change

mitigation in the water sector. The collaboration with local research collaborators and government officials has allowed us to identify key issues and priorities for water management in the studied river basins. In the case of Cambodia and Bangladesh where the first two workshops were held, the lack of wastewater treatment facilities and associated river pollution represented the foremost concern and policy priority for local water authorities. Downstream ecosystem impacts and GHG emissions have not been incorporated into any pro-active policy alternatives. To compare the poor wastewater infrastructure in the Mekong and Ganges basins with growing environmental awareness and technological development in China, the third workshop included discussion and interviews with local watershed managers from the YRCC. Based on these discussion rounds and critical examinations of the current water policies, policy recommendations for addressing water pollution impacts on riverine ecosystem process and GHG emissions will be summarized in the form of a policy brief or brochure. These policy recommendations, together with project findings translated in language intelligible to the general public, will be disseminated via the project homepage and brochures to collaborating local water agencies and NGOs.

4. Conclusions

This project aimed to build a collaborative research network to assess the current status of water pollution impacts on C export and GHG emissions from Asian rivers, using monitoring data available from literature and local sources and new field measurements from fieldworks conducted by project members. Specifically, three workshops were held in a regional hub of each of three studied rivers (the Mekong, the Ganges, and the Yellow River) to bring together researchers working on Asian river systems across the Asia-Pacific region to assess the current status of human impacts on riverine C fluxes and GHG emissions. As part of the workshops and follow-up field studies, collaborative research activities involving project members and other local collaborators were implemented through interrelated project components, including reviewing literature, synthesis of available monitoring data and new field measurements, and establishing and disseminating standardized riverine C monitoring protocols.

Comparing literature information and local data with new field measurements suggests that rising organic pollution in rivers and tributaries downstream of metropolitan areas across the region has greatly increased organic C and GHG fluxes over the recent decades. While rising organic pollution resulting from rapid urbanization and poor wastewater management is adding additional perturbations to impoundment-affected reaches across the Mekong and Yellow River, the problem was particularly severe in the most densely populated Ganges basin, as illustrated by extremely high concentrations of organic C and GHGs along the Yamuna traversing the Delhi metropolitan area. Combining model-based estimation with field measurements of $p\text{CO}_2$ provided a more reliable, regional-scale estimate of riverine CO_2 emission from the Indian Subcontinent, which reflected recent increases in organic loads in wastewater from urbanizing rivers across the region. These results call for concerted efforts among scientists and water agencies that can properly assess water pollution impacts on regional and global river carbon budgets and explore alternative policies for mitigating wastewater-induced increases in GHG emissions.

We recommend alternative policies integrating the conventional water quality focus and a new initiative for mitigating riverine GHG emissions. Literature reviews and our field measurements emphasized urban wastewater and downstream rivers as hotspots for GHG

emissions from urbanizing river basins, suggesting that policymakers and watershed managers set priority areas and target concentration ranges of riverine GHGs across urban river systems to mitigate wastewater-enhanced GHG emissions from the water section. Outcomes of this project, particularly standardized monitoring protocols and the growing collaboration network among researchers with expertise on river C fluxes, can contribute to capacity building and scientific information required for policymakers and water agencies to envisage proactive water policies mitigating GHG emissions from rapidly urbanizing river systems across Asia and worldwide.

5. Future Directions

Building on the collaboration network including the project members and invited speakers of the three workshops, follow-up research activities can expand the duration of the ongoing collaboration and the scope of addressed research topics. To add up to the first basin-wide field surveys of three GHGs along the Ganges, project members (Drs. Kumar and Tareq), in collaboration with PI, will continue field monitoring in India and Bangladesh, following monitoring protocols established by this project. Dr. Tareq, in collaboration with PI, acquired a research grant from the International Environmental Research Institute (IERI), GIST, Korea (entitled “Impact of climate changes on water quality and greenhouse gas emission of the Ganges-Brahmaputra River, Bangladesh”; 10,000 USD for Jan 1–Dec 31, 2019) In situ incubation experiments and continuous sensor measurements of $p\text{CO}_2$, which were successfully implemented in the Mekong and Ganges, can also be applied to the Yellow River by project members (Drs. Lu, Ran and Yu) with technical support from PI.

An ad-hoc review team, involving project members (Drs. Chae, Lu, Richey and Xuan) and workshop invited speakers (Dr Andreas Lorke, University of Koblenz, Germany and Dr Qiuwen Chen, Nanjing Hydraulic Research Institute) working on GHG emissions from the Mekong, will be formed to review emerging issues on altered river flow and C fluxes by newly constructed dams and aggravating water pollution in the Mekong River. While this project focused on the role of polluted urban rivers as sources of GHGs, a follow-up project can be envisaged to explore transitional stages of eutrophic rivers from a C sink, where phytoplanktonic CO_2 uptake exceeds CO_2 released from OM biodegradation to a source of GHGs, where emissions of CO_2 and other GHGs overwhelm temporary or seasonal increases in phytoplanktonic CO_2 uptake. These future activities adding to the project’s focus on Asian river systems will contribute to elucidating human impacts on global riverine budgets of CO_2 and other GHGs.

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7. Appendix

List of participants from three APN workshops

The First International Workshop on Human Impacts on Carbon Fluxes in Asian River
Systems, January 11–13, 2017
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The Third International Workshop on Human Impacts on Carbon Fluxes in Asian River
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Inner Mongolia University, Hohhot, China

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<Articles published in international journals>

- **Biogeosciences Special Issue: Human impacts on carbon fluxes in Asian river systems**

- Editor(s): **J.-H. Park, V. V. S. S. Sarma**, G. Abril, and D. Butman
- List of papers published by project members (project members indicated in bold font)

Park, J.-H., Nayna, O. K., Begum, M. S., Chea, E., et al. (2018). Reviews and syntheses: Anthropogenic perturbations to carbon fluxes in Asian river systems – concepts, emerging trends, and research challenges. *Biogeosciences*, 15(9), 3049–3069. doi: 10.5194/bg-15-3049-2018

Ran, L., Tian, M., Fang, N., Wang, S., **Lu, X.**, Yang, X., & Cho, F. (2018). Riverine carbon export in the arid to semiarid Wuding River catchment on the Chinese Loess Plateau. *Biogeosciences*, 15(12), 3857–3871. doi: 10.5194/bg-15-3857-2018

Le, T. P. Q., Marchand, C., Ho, C. T., Duong, T. T., Nguyen, H. T. M., **Lu, X. X.**, ... Le, N. D. (2018). CO₂ partial pressure and CO₂ emissions from the lower Red River (Vietnam). *Biogeosciences*, 15, 4799–4814, doi: 10.5194/bg-15-4799-2018

Dutta, M. K., **Kumar, S.**, Mukherjee, R., Sanyal, P., & Mukhopadhyay, S. K. (2019). The post-monsoon carbon biogeochemistry of the Hooghly–Sundarbans estuarine system under different levels of anthropogenic impacts. *Biogeosciences*, 16(2), 289–307. doi: 10.5194/bg-16-289-2019

Lin, L., **Lu, X.**, Liu, S., Liang, S.-Y., & Fu, K. (2019). Physically controlled CO₂ effluxes from a reservoir surface in the upper Mekong River Basin: a case study in the Gongguoqiao Reservoir. *Biogeosciences*, 16(10), 2205–2219. doi: 10.5194/bg-16-2205-2019

Jin, H., Yoon, T. K., **Begum, M. S.**, Lee, E.-J., Oh, N.-H., Kang, N., & **Park, J.-H.** (2018). Longitudinal discontinuities in riverine greenhouse gas dynamics generated by dams and urban wastewater. *Biogeosciences*, 15(20), 6349–6369. doi: 10.5194/bg-15-6349-2018

Li, S., Mao, R., Ma, Y., & **Sarma, V. V. S. S.** (2019). Gas transfer velocities of CO₂ in subtropical monsoonal climate streams and small rivers. *Biogeosciences*, 16(3), 681–693. doi: 10.5194/bg-16-681-2019

Krishna, M. S., Viswanadham, R., Prasad, M. H. K., Kumari, V. R., & **Sarma, V. V. S. S.** (2019). Export fluxes of dissolved inorganic carbon to the northern Indian Ocean from the Indian monsoonal rivers. *Biogeosciences*, 16(2), 505–519. doi: 10.5194/bg-16-505-2019

- **Journal articles in review or preparation**

–**Park, J.-H., Nayna, O. K.**, Abril, G., Alcamo, J., **Begum, M. S.**, Butman, D., Flörke, M., & **Sarma, V. V. S. S.** Where is all the carbon in wastewater going? (Submitted to *Science* on 1 August 2019)

–**Nayna, O. K., Begum, M. S., Sarma, V. V. S. S., Kumar, S., Tareq, S. M.**, Butman, D., & **Park, J.-H.** Anthropogenic impacts on riverine carbon fluxes – Revised estimates of carbon dioxide emission from the Indian subcontinent.

–**Nayna, O. K., Begum, M. S., Park, J.-H., et al.** Improving the estimation of *p*CO₂ from carbonate equilibria (pH and alkalinity) in large polluted Asian rivers.

–**Nayna, O. K., Begum, M. S., Chea, E., Xuan, D. T., Tareq, S. M., Sarma, V. V. S. S., Kumar, S., Lu, X., Ran, L., Yu, R., & Park, J.-H.** Regional-scale assessment of organic pollution impacts on riverine C fluxes and GHG emissions.

- Begum, M. S.**, Jang, I., Oh, H. B., Jin, H., **Nayna, O. K.**, Chun, Y., Haque, M., **Tareq, S. M.**, & **Park, J.-H.** Enhanced riverine GHG evasion associated with altered OM from urban pollution in the Ganges river system.
- **Begum, M. S.**, **Nayna, O. K.**, Lee, Y.-K., Hur, J., Bogard, M., Butman, D., **Chea, E.**, **Xuan, D. T.**, **Tareq, S. M.**, **Lu, X.**, **Ran, L.**, **Yu, R.**, & **Park, J.-H.** GHGs emission from pollution prone large Asian Rivers controlled by dissolved organic matter flux and composition.
- Jin, H., **Begum, M. S.**, Yoon, T. K., **Chae, E.**, Sang, Z., Aing, C., Oh, N.-W., Lee, E.-J., & **Park, J.-H.** Urban wastewater as a source of riverine greenhouse gas outgassing from the Mekong–Tonle Sap river system.
- Park, J.-H.**, **Chae, E.**, Chen, Q., Lorke, A., **Lu, X.**, **Richey, J. E.**, & **Xuan, D. T.** A review of impoundment impacts on carbon fluxes in the Mekong River.

<Conference paper/presentations>

- **First workshop abstracts** (refer to uploaded files on the project webpage; <http://apn.peblab.com>)

<p>Ji-Hyung Park Water Pollution Impacts on Carbon Export and Greenhouse Gas Evasion from Asian River Systems – Project Overview</p>
<p>David Butman, Rob Striegl, Sarah Stackpoole, Paul del Giorgio, Yves Prairie, Darren Pilcher, Peter Raymond, Fernando Paz Pellat, Javier Alcocer A National Scale Assessment of Carbon Fluxes from Inland Waters of the US in the Context of the 2nd U.S. State of the Carbon Cycle Report</p>
<p>Gordon W. Holtgrieve, Benjamin Miller, Mauricio E. Arias, John Sabo, CHHENG Phen, Vittoria Elliott, SO Nam Flood-pulse Controls on Carbon Fluxes and Fishery Productivity in the Mekong River and Tonle Sap Lake</p>
<p>Xi Xi Lu, Ran Lishan, Yu Ruihong, Hu Haizhu Carbon Outgassing from the Yellow River: An Overview</p>
<p>Lishan Ran and X.X. Lu Riverine CO₂ Emissions from the Wuding River Catchment on the Loess Plateau: Environmental Controls and Dam Impoundment Impact</p>
<p>Ruihong Yu, Xixi Lu, Haizhu Hu, Lishan Ran, Lingyu Li, Mingyang Tian Spatio-temporal Variations and Influencing Factors of Carbon Dioxide Evasion from the Yellow River: An Example of the Toudaoguai Station</p>
<p>VVSS Sarma Trace Gases Emissions from the Indian Estuaries</p>
<p>Shafi M Tareq, Sharmin Y Rikta, M. Shiblur Rahman Using 3DEEM Fluorescence Spectroscopy for Monitoring Water Quality and DOM Dynamics in the Ganges and Brahmaputra River in Bangladesh</p>
<p>Adrienne P. Smits, Simone R. Alin, Jeffrey E. Richey, Gordon W. Holtgrieve Seasonal Dynamics of the Sources and Isotopic Composition of Dissolved Inorganic Carbon in Large Tropical River Systems: the Mekong and Chao Phraya Rivers of Southeast Asia</p>
<p>Eliyan Chea Current Water Quality at Mekong River</p>
<p>Do Thi Xuan Dynamic Changes of Quality of Surface Water in the Mekong River in Vietnam from 2013–2015</p>
<p>Tae Kyung Yoon, Hyojin Jin, Most Shirina Begum, Ji-Hyung Park Water-air CO₂ Flux Measurement Methods: Principles, Validations, and Challenges</p>
<p>Most Shirina Begum, Tae Kyung Yoon, Ji-Hyung Park Sampling and Analysis Protocols for Assessing Human Impacts on the Riverine Carbon Fluxes in Asian River Systems</p>
<p>Tae Kyung Yoon, Hyojin Jin, Most Shirina Begum, Ji-Hyung Park Project Protocol</p>

- **Second workshop abstracts** (refer to uploaded files on the project webpage; <http://apn.peblab.com>)

<p>Ji-Hyung Park Assessing Pollution Impacts on Carbon Fluxes in Asian River Systems – Workshop Outcomes, Exploratory Field Studies, and Data Synthesis</p>
<p>Tarun K. Dalai Enhanced Delivery of Dissolved Inorganic Carbon from the Ganga (Hooghly) River Estuary to the Bay of Bengal: Sources, Processes and Fluxes</p>
<p>VVSS Sarma, M.S. Krishna, R. Viswanadham, M.H.K. Prasad Export Fluxes of Dissolved Inorganic Carbon to the Northern Indian Ocean from the Indian Monsoonal Rivers</p>
<p>Sanjeev Kumar Effect of a Tidal Cycle on Biogeochemistry of Mangrove Dominated Tropical Estuary (Sundarbans, India)</p>
<p>Shafi M. Tareq Characteristic and Dynamic of Dissolved Organic Matter in the Ganges and Brahmaputra River, Bangladesh</p>
<p>Xi Xi Lu, Yang Xiankun, Tian Minyang, Su Yuanrong, Ran Lishan, Hu Haizhu, Yu Ruihong Carbon Emission from Tibet Plateau Rivers: A Case Study of the Yellow River Headwater Region</p>
<p>Lishan Ran, Mingyang Tian, Xi Xi Lu Assessing Riverine Carbon Budget in the Wuding River Basin on the Loess Plateau</p>
<p>Eliyan Chea Phnom Penh Wastewater Management and Pressures to River Water Quality</p>
<p>Most Shirina Begum, Hyojin Jin, Ji-Hyung Park Organic Matter and Greenhouse Gas Dynamics in Three Asian River Systems – Monitoring Protocol and Preliminary Results</p>
<p>Omme Kulsum Nayna, Yewon Chun, Ji-Hyung Park Multiple Approaches to Improving Accuracy of Estimating Greenhouse Gas Emission from Asian River Systems</p>
<p>Most Shirina Begum, Hyojin Jin, Tae Kyung Yoon, Ji-Hyung Park Project Protocol</p>

- **Third workshop abstracts**

<p>Ji-Hyung Park Synthesis of Project Outcomes toward a Regional-Scale Assessment of Water Pollution Impacts on Greenhouse Gas Emissions from Asian River Systems</p>
<p>Zaihua Liu Carbon sink by coupled carbonate weathering with aquatic photosynthesis—Control of climate and land-use changes</p>
<p>Andreas Lorke, L. Liu, Z.J. Yang, K. Delwiche, L.H. Long, J. Liu, S.B. Xiao, D.F. Liu, C.F. Wang, P. Bodmer, and L.I. Steinle Spatial and temporal variability of methane emission from cascading reservoirs at the Upper Mekong River</p>
<p>Mingming Hu, Yufei Bao, Pengcheng Du, Aimin Cai, Cong Lv, Shanze Li, and Yuchun Wang Export fluxes of dissolved inorganic carbon to the Northern Indian Ocean from the Indian monsoonal rivers</p>
<p>Sanjeev Kumar Nitrogen and carbon cycling study in terrestrial aquatic and forested ecosystems in India</p>
<p>Shafi M. Tareq, Morshedul Haque, Nahin Mostofa Niloy, and M Shiblur Rahaman Investigating water quality of the Ganges and Brahmaputra river using three dimensional excitation emission matrix (3DEEM) fluorescence spectroscopy</p>
<p>Haizhu Hu, Xi Xi Lu, Xiankun Yang, Lishan Ran, Chen Gong, Mingyang Tian, Yuanrong Su, Ruihong Yu Carbon dioxide emission from surface water in cascade reservoirs of the Yellow River source region</p>
<p>Xi Xi Lu, Ruihong Yu, Haizhu Hu, and Lishan Ran Carbon emissions from China's waterbodies: an overview</p>
<p>Yuanrong Su, Ruihong Yu, Xi Xi Lu, Lishan Ran, Xiankun Yang, Haizhu Hu, and Zhuangzhuang Zhang Temporal and spatial variation of major ion chemistry and dissolved carbon in the source region of the Yellow River</p>
<p>Lishan Ran and Hongyan Shi Hydrological controls on riverine export of water, sediment, and carbon: a case study of the Wuding River on the Chinese Loess Plateau</p>
<p>Most S. Begum, Omme K. Nayna, Hyojin Jin, and Ji-Hyung Park Assessing Greenhouse Gas and Organic Matter Flux and Quality in Urbanized Asian Rivers</p>
<p>Omme K. Nayna and Ji-Hyung Park Three approaches to assessing pollution impacts on riverine organic carbon load and greenhouse gas emissions</p>
<p>Most Shirina Begum, Hyojin Jin, Tae Kyung Yoon, and Ji-Hyung Park Project protocol</p>

- **Other conference abstracts**

1) Presented at International Society of Limnology XXXIV Congress 2018

Ji-Hyung Park, Most Shirina Begum, Omme K. Nayna, Hyojin Jin, Yewon Chun

Longitudinal Discontinuities in Riverine CO₂ Dynamics in Urbanizing Asian River Systems

Abstract – CO₂ emissions from Asian river systems represent a critical missing piece of information required for an accurate estimation of the global riverine C budget. Rapid urbanization across major Asian river basins may affect riverine metabolic processes and CO₂ emissions, adding a large uncertainty to C budget estimates. Literature survey was combined with explorative field measurements in three large Asian rivers (the Ganges, Mekong, and Yellow River) to examine how urbanization and associated water pollution affect the surface water partial pressure of CO₂ (pCO₂) via altered riverine metabolic processes along the hypothetical river continuum, which has long been used to explain longitudinal variations in “natural” riverine processes. In many Asian rivers including the three monitored rivers, the level of pCO₂ was much higher along the lower reaches and tributaries downstream of large metropolitan areas, compared to relatively low pCO₂ values observed in headwater systems. The up- and downstream comparison of dissolved organic matter (DOM) characteristics, including dissolved organic carbon (DOC) concentrations and stable isotope ratios, DOM optical intensities, and molecular signatures identified by ultrahigh resolution mass spectrometry, commonly pointed to significant alterations of the DOM composition in the affected reaches by DOM moieties derived from urban wastewater and tributaries. In-stream incubation experiments conducted with river and polluted tributary waters in the Ganges (near Dhaka) and Mekong (along Phnom Penh) revealed boosting effects of anthropogenic and planktonic DOM moieties on the biodegradation of DOM and CO₂ emission. The results suggest that high levels of water pollution in rapidly urbanizing river systems can shift the balance of autotrophy and heterotrophy in eutrophic river reaches toward a high C-leaking state that deviates from the gradual longitudinal pattern envisioned by the traditional river continuum concept.

2) Presented at Korean Society of Limnology Feb 2019

Most Shirina Begum, Omme Kulsum Nayna, Ji-Hyung Park

Anthropogenic alteration of dissolved organic matter characteristics resulting in large greenhouse gas emissions from highly urbanized Asian rivers

Abstract—Large uncertainties remain in estimating global riverine emissions of CO₂ and other greenhouse gases (GHGs) due to lack of field measurements especially in pollution prone Asian rivers. To explore how urbanization and associated water pollution affects riverine metabolic processes and GHG emissions, two seasonal field surveys were conducted in three large Asian Rivers (the Ganges, Mekong and Yellow River) as part of an international

collaborative research. Measurements of surface water concentrations of the CO₂, CH₄ and N₂O along the mainstem and polluted local tributaries of the three rivers revealed distinct longitudinal patterns of generally increasing gas concentrations along downstream reaches and tributaries, but the levels of pCO₂ were persistently lower across the eutrophic impounded reaches. Concomitant measurement of riverine dissolved organic carbon (DOC) concentration showed similar pattern of downstream increase in these river systems. Optical and molecular characterization of dissolved organic matter (DOM) revealed higher amount of protein-like fluorescent DOM and abundance of N and S containing compounds at the downstream reaches and urban tributaries of Ganges suggesting influence of urban pollution. In-situ incubation at a downstream location of Ganges further elucidated enhanced biodegradability and higher CO₂ emission upon mixing with polluted water from urban tributary. The higher GHG levels and altered DOM properties along the tributaries and downstream reaches of the three rivers suggest that rising water pollution in rapidly urbanizing Asian rivers are altering not only material fluxes but also riverine metabolic processes leading to large increases in GHG emissions.

Funding sources outside the APN

Support Leveraged from sources other than APN: Budget Secured from Other Sources (Cash and In-kind Contribution)		In-Kind (US\$)	Cash (US\$)
Activity	Organisation		
Establishing C monitoring protocols (Project component 2): instruments (5 CO ₂ sensors, 1 CO ₂ analyzer, 1 Gas Chromatograph); consumable cost provided by PI project (5/2015-4/2016; total funding: 90,000 US\$)	Ewha Womans University	70,000	5,000
Field study (Project component 3): instrumental support (2 CO ₂ analyzers) and sampling cost provided from National University of Singapore-funded project (7/2015-6/2018) for field study in China	National University of Singapore	20,000	24,000
Personnel support (research fellow monthly salary or scholarship for a Ph.D. student for 3 years)	Ewha Womans University		35,000
Total		90,000	64,000

List of Young Scientists

□ **Tae-Kyung Yoon**

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- *Involvement in the project*

Dr. Yoon worked as a post-doctoral researcher in the first year of the APN project since 2016 to 2017. He worked on developing the monitoring protocol and provided a hand-on demonstration during the field work at first APN workshop in Phnom Penh, Cambodia in January 2017. Within his short time of involvement with APN, he was great help in planning and executing the field works in the first year.

- *Short message for APN*

“Even though I worked for the project only for a short period of time, the novel, international collaboration supported by the APN was remarkable for me to enhance my career, knowledge, and experiences. I hope further collaboration among the participants continues to keep dealing with the urgent climate change issues in Asia-Pacific societies.”

□ **Most Shirina Begum**

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- *Involved in the project as a Ph.D.*

Ms. Begum has worked as a research assistant for the APN project since 2017 to 2019. However, she was involved with the project since the beginning as project coordinator under the Project leader’s guidance. She was involved in planning and executing field work, i.e., sample collection, field experiment, and cruise underway measurements in the Ganges, Mekong and Yellow Rivers, as well as laboratory experiments and analyses. During the three workshops, Begum not only presented the protocol (developed by project leader’s team) and key results but also helped the hosts of the workshop with logistics, reimbursement, and proceedings. Finally, she prepared the annual reports (and financial) and the manuscript for the APN Science Bulletin together with other members under the project leader’s direct supervision.

- *Short message for APN*

“I was working on the APN project since the beginning of my PhD, and was actively involved with all the research and administrative work. I have learnt a lot from working for the project, and I got to meet with a number of renowned researcher in my field during the workshops. At times it was difficult to follow up with all the laboratory works, reporting, and other administrative work; however, it helped me develop multi-tasking skills which is important in

academia. I hope this experience will help me to become a successful and productive researcher in future. I would like to express my sincere gratitude to APN for the opportunity and I hope to keep in touch with APN for future grants, conference and collaboration.”

□ **Omme Kulsum Nayna**

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- *Involved in the project as a Ph.D. student (scholarship provided by Ewha Womans University)*

Ms. Nayna was involved with the project since 2017 to 2019 as a research assistant. She actively participated in the field sampling and laboratory analysis under the project leader's guidance. She also collected data from various global database, local monitoring organization and existing literature in order to develop a regional budget of CO₂ and other GHGs. She worked on developing and improving multiple approaches to estimate riverine CO₂ and other GHGs along with riverine OM as DOC and TOC. Nayna actively participated in publishing the data in forms of journal article, conference presentation including two of the APN-funded workshops. She helped project leader in preparing reports and publishing the outcomes of the project.

- *Short message for APN*

“I was working on the APN project since the beginning of my PhD from 2017 to present and I actively participated in all the researches and two workshops (Dhaka and Hohhot). It was an excellent experience for me to meet with renowned scientists in my field during the workshops and share the research ideas. I learnt a lot from the field sampling and experiments, cruise underway measurements, laboratory analyses, data synthesis and publishing article during the working period of this project. I learnt how to collaborate with other researchers during the project work, I hope all these experiences will help me to be a successful researcher in this field in future. I would like to thank APN for my valuable experiences and I hope to keep in touch with APN in future for research grants, collaboration and conference.”

□ **Md. Morshedul Haque**

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- *Involved in the project as a MSc student of Jahangirnagar University*

Mr. Haque was involved with the project since 2017 to 2019 and contributed to various project activities. Besides helping with collecting samples from Ganges in Bangladesh, Shamim conducted the monthly C monitoring in Ganges under supervision of Dr. Tareq (project member) for 1 year. Shamim helped Dr. Tareq in organizing the second workshop in Dhaka, Bangladesh in Feb 2018 and assisted with the field trip. He will be involved in publishing the APN outcomes from the follow up Ganges monitoring with the Project leader's

team.

- *Short message for APN*

“Firstly, I am very glad to have the opportunity to work on the APN–funded project. I learned a lot of basic and innovative knowledge of research through this project. Basically, the technical skills that I gained from the project workshop and the monthly C monitoring in Ganges has inspired me to work more on this field. I would like to thank APN for my valuable experience and wish APN will encourage more young people around the world to embark of their project.”

□ **Hyojin Jin**

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- *Involved in the project as a Ph.D. student of Ewha Womans University*

Ms. Jin was involved with two of the APN workshops and helped with sampling and other field works. In particular, she helped with the underway cruise measurement of $p\text{CO}_2$ and other water quality parameters with technical her technical skills on CO_2 sensor during the field trips in first and second APN workshops.

□ **Yewon Chun**

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- *Involved in the project as a MSc. student of Ewha Womans University*

Ms. Chun was involved with two of the APN workshops and helped with sampling and other field works. In particular, she helped with the underway cruise measurement of $p\text{CO}_2$ and other water quality parameters with technical help on CO_2 measurements with equilibrators systems during the field trip in second APN workshop.

Glossary of Terms

Acronyms/Abbreviations	Description
C	Carbon
OM	Organic matter
DOM	Dissolved organic matter
DOC	Dissolved organic carbon
BDOC	Biodegradable dissolved organic carbon
POC	Particulate organic carbon
TOC	Total organic carbon
TOW	Total organics in wastewater
DIC	Dissolved inorganic carbon
BOD	Biochemical oxygen demand
COD	Chemical oxygen demand
DO	Dissolved oxygen
EC	Electrical conductivity
TA	Total alkalinity
GHG	Greenhouse gas
$p\text{CO}_2$	Partial pressure of CO_2
GC	Gas chromatography
FID	Flame ionization detector (for CO_2 and CH_4 analysis)
uECD	Micro electron capture detector (for N_2O analysis)
EEM	Excitation-emission matrix
PARAFAC	Parallel factor analysis
FI	Fluorescence index
HIX	Humification index
BIX	Biological index
UVA	Ultraviolet absorbance
SUVA ₂₅₄	Specific ultraviolet absorbance at 254 nm
FDOM	Fluorescent dissolved organic matter
C1	Humic-like FDOM
C2	Microbial humic-like FDOM
C3	Protein-like FDOM
PCA	Principal component analysis
SPE	Solid Phase Extraction
FT-ICR-MS	Fourier transform ion cyclotron resonance mass spectrometry
ESI	Electro spray ionization
WWTP	Wastewater treatment plant
TSS	Total suspended solid
GF/F	Glass fiber filter
PTFE	Politetrafluoroethane
CO2SYS	CO_2 system
IRGA	Infrared gas analyser
GLORICH	Global river chemistry database
CPCB	Central pollution control board
IPCC	Intergovernmental panel on climate change